# ISOTOPES AND RADIATION TECHNOLOGY

*A Quarterly Technical Progress Review*

Prepared for DIVISION OF TECHNICAL INFORMATION / DIVISION OF ISOTOPES DEVELOPMENT, UNITED STATES ATOMIC ENERGY COMMISSION, by P. S. BAKER, A. F. RUPP, and ASSOCIATES, OAK RIDGE NATIONAL LABORATORY

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**FEATURE ARTICLES:**

118 Medical Radioisotope Cows

# TECHNICAL PROGRESS REVIEWS

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# ISOTOPES AND RADIATION TECHNOLOGY

## A REVIEW OF RECENT DEVELOPMENTS

Prepared for DIVISION OF TECHNICAL INFORMATION / DIVISION OF ISOTOPES DEVELOPMENT, UNITED STATES ATOMIC ENERGY COMMISSION, by P. S. BAKER, A. F. RUPP, and ASSOCIATES, OAK RIDGE NATIONAL LABORATORY

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# Foreword

*Isotopes and Radiation Technology* is a Technical Progress Review prepared by the Isotopes Development Center, Oak Ridge National Laboratory, at the request of the Divisions of Technical Information and Isotopes Development, U. S. Atomic Energy Commission. This Review is intended to assist those interested in keeping abreast of significant developments in the fields of isotopes and radiation technology. It is not a comprehensive review of all literature published in this field during a given quarter; rather, it is a mechanism for presenting concise, selected reviews of information on subjects of prevailing Commission interest as it becomes available.

This Review attempts to relate the published results of research and development sponsored by the Division of Isotopes Development to significant developments in radioisotopes and radiation technology, as reported in the world literature. Coverage includes isotope production and development, isotope technology development, isotope applications technology, process radiation development, and radiation processing of food and medical supplies.

In addition to reviews of current literature and special review articles dealing with specific isotopes, facilities, and applications, this publication occasionally contains feature articles prepared by recognized experts on specific topics of current interest. Critical evaluations and interpretations presented are those of the editors and invited reviewers; therefore readers are encouraged to consult the original references in order to obtain all the background of the work reported and the interpretation of the results given by the original authors.

The subject headings listed below have been adopted to help maintain continuity and order, from one issue to the next, in the material reviewed. All reviews, except Feature Articles, will be arranged under these headings; but any one issue will not necessarily contain all the headings.

Isotope Production and Development
Isotope Technology Development
Food Irradiation
Process Radiation Development
Source Development
Isotopic Power Development
Applications in Medicine
Safety
Isotopes Around the World
Miscellaneous Items of Interest

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FEATURE ARTICLE

*Radioisotope "cows" may be "milked" in industrial work as well as in hospitals, and this article should be of interest to anyone who is considering the use of short-lived radioisotopes. It is the product of a 5-year study by the author.*

# 118 Medical Radioisotope Cows

**By Marshall Brucer, M. D.***

*The production of short-lived radioisotopes from cows makes it possible to use these materials in places remote from a reactor or an accelerator. Of 118 radioisotope cows that are available, 8 are practical because of medical necessity or physical availability, 11 may be practical because of their short half-lives, and 16 more could be made available if the need for them became urgent. Originally a radioisotope cow was a related set of isotopes in which the parent decayed through a daughter to a stable isotope or had a half-life longer than its daughter; the daughter was a different element and hence could be "milked," or a generator could be developed for easy milking. The idea has now been extended to include both "isomer" and "reverse" cows. The first radioisotope produced by a cow was $^{132}I$, milked from a $^{132}Te$ parent.† A few others have been suggested and a few generators have been developed. To make a generator is a sophisticated technical task, which may make unusual reactor or cyclotron demands. But the practicality of a medical radioisotope cow should not be a problem of chemical feasibility; rather it is a problem of medical necessity.*

A radioisotope cow is not bovine, though some librarian is going to file this article under "Agriculture — Fallout In." The article has nine tables. Almost every mark on each table is open to argument. There will be trivial arguments on physical details: not trivial to a physicist or chemist, but trivial to a physician. This is especially true since I will accept the 1961 Sullivan Chart of Nuclides‡[1] as absolutely true. It isn't absolutely true, but it is adequate for the purpose of looking at medical radioisotope cows.

Far more serious are the arguments that will be generated by such decisions as "lead has a medical use, but cadmium doesn't." No toxicologist can accept this. There is a built-in argument when I say that 22-hr $^{43}K$ is short-lived, and therefore we must have 12-hr $^{42}K$; but a 12-hr isotope available to a physician on demand is long-lived compared to a 22-hr isotope in the Oak Ridge catalog. I know that $^{42}Ar$ is usually a gas hard to control, but this is a worry for the Isotopes Development Center, not for physicians who want to do tumor detection with $^{42}K$.

If the Chart of Nuclides were a matter of design rather than circumstance, we would obviously design an oxygen activity that could be turned on and off at will. Since this cannot be done, we are forced to compromise. The "discovery" of new isotopes that happen to *almost* fit a need was a legitimate exercise when radioisotopes were new and needed a good press. But now they have to earn their keep, and the older principles of pharmacology are reasserted. If we are studying potassium, we must use potassium; rubidium is a second-rate substitute. It is just as true that in *some* potassium

*In May 1965 Dr. Marshall Brucer received the U. S. Atomic Energy Commission citation for outstanding service to the nation's nuclear energy program. At that time a colleague commented on his writing: "His irreverent and unconventional approaches often are a greatly needed breath of fresh air." At the time of his retirement in 1962 from Oak Ridge Institute of Nuclear Studies, Dr. Brucer was head of its Medical Division, which he organized in 1949. He now lives in Tucson, Ariz., and continues to write prolifically with interesting and provocative Brucerian phraseology.

†In Dr. Brucer's original manuscript, both atomic number and atomic weight were shown for each isotope. The atomic numbers have been deleted in the text but have been left in the tables.

‡Revisions of chart up through January 1961 were used in this article.

studies 19-day rubidium is pharmacologically better than half-day potassium. Occasionally the pharmacology of an element doesn't matter.

## Changing Concept of Half-Life

Half-life measured in terms of the sun's coming up and going down (my observation, not an astronomer's) is not going to change much for many radioisotopes. But the half-life practicality in terms of physician–patient–hospital red tape is in violent flux. Diagnostic tests are not chronologic in terms of the amount of time an isotope spends inside the body. The time it takes to perform a diagnostic test is more truthfully measured as: time to transport the patient from bed to laboratory, the current state of health of the hospital procurement officer, the physician's interest, the laboratory's efficiency, and a hundred other details of hospital procedure. Hospitals work on a "day" time scale. An operating room may work on an "hour" time scale, but the human body seldom works on a "minute" time scale.

The logistics of laboratory–hospital–patient–physician scheduling make the full day an attractive time-marker. "About-a-week" became the "medium half-life" during the late 1930's when Hamilton found it difficult to do thyroid metabolic studies with 25-min $^{128}I$ (the only cyclotron-produced iodine then available).[2] The resurgence of "steady-state" studies in pharmacology during the 1930's put a premium on long-lived radioisotopes.

A few years ago 2.7-day $^{198}Au$ had an impossibly short life. Improvements in commercial production and delivery have changed "impossibility" to "nuisance," but there are still some time limits beyond argument. A daughter radioisotope that has a microsecond half-life may be enormously important in the theory of physical decay but not in the human body.

I am going to arbitrarily class any daughter with a half-life of seconds as a part of the parent and not as a separate isotope. This arbitrary decree could probably be pushed into the minute range for most clinical uses, but the cesium–barium cow already has a usefulness (technologically clinical, not clinically clinical), and this might be going too far. There are very rapid biological conversions, especially in neurology, but these haven't been studied with radioisotopes yet.

The same kind of decision could be made with very long-lived isotopes. A geologist may consider a million years a short time, but my life-span is less than 100 years, and to me a century is longer than life. For the purpose of talking about radioisotope cows in clinical medicine, I can make some definitions, although the rules are not rigid:

Biologically stable: half-life greater than a million years ($^{40}K$ is an exception)
Very long lived: half-life longer than a hundred years (usually medically useless; $^{14}C$, $^{36}Cl$, $^{226}Ra$, and a number of others are exceptions)
Long lived: half-life one to a hundred years
Medium lived: half-life greater than a week but less than a year (usually useful medical radioisotopes; only about twenty are used now, but there promise to be many)
Short lived: half-life greater than 24 hr but less than a week (usually pharmacologically questionable)
Very short lived: half-life a minute to a day (somewhat difficult to work with medically; $^{18}F$, $^{99m}Tc$, and $^{24}Na$ are medically desirable exceptions)
Biologically instantaneous: half-life less than 60 sec

It is intellectually dangerous to consider these definitions as more than a tentative oversimplification, and I repeat that the rules cannot be rigid.

It would be desirable to make similar definitions of energy, range, chemical reactivity, and biological usefulness, but these are not immediately at issue. Radioactive cows are first of all a matter of half-life. The idea of separating isotopes by half-life was implicit in the 1898 Rutherford–Owens studies[3] of thoron, even before the idea of an isotope was developed. But the 1951 Winsche–Stang–Tucker description of the $^{132}I$ generator was the beginning of the exploitation of a half-life decay differential between parent and daughter to capitalize on the occasional clinical desirability of short life.[4,5]

## Different Breeds of Cows

Many clinical tests demand only minutes of radioactivity, e.g., circulation times, cardiac outputs, and some initial turnovers. Some tumor uptakes are maximum in hours and useless beyond this time. When the radioisotope cow was first developed, radioisotopes still

were little understood and a short life meant a lower dose. A few years later the low-dose argument was used for $^{125}I$: the *longer* half-life gave a lower dose. If the radiation hysteria hadn't been such a deterrent to medical use, the obvious inconsistency would have been amusing.

Another consideration is of greater importance. The cow was originally conceived to be used clinically, but the manufacturer of the chemical drug has an advantage in working with a short life that decays to a longer life—the reverse of the cow idea. The pharmacologist hasn't yet studied the unique advantages of a drug that injects itself, not intravenously or subcutaneously or orally, but from the site of its metabolism. This is the "reverse cow."

Shortly after $^{125}I$ became available, I. M. Ariel in New York began using a combination $^{125}I$–$^{131}I$. The short life was for therapy, the long life was for simultaneous diagnosis of the results of therapy. Both the differences and the similarities of biologic behavior are intriguing. More important to isotopologists, similar isomer pairs are potential diagnostic tools, both short-to-long and long-to-short; these are isomer cows.

The 1951 Brookhaven cow was not the isolated animal we once thought ingenious. It was the beginning of a herd of about 118 (or maybe more) cows. There are short cows, long cows, reverse cows, isomer cows, reverse isomer cows, and many combinations. Since chemistry is still part of the clinical laboratory, even the stable isotope from a decay chain might be useful.

Before attempting a judgment of what is useful or desirable, we need to know what is possible.[6,7] I blithely assume that the isotopes development people can make almost anything, provided it at least exists. The crass engineering details I leave to them.

## *Radioisotope Cows That Are Practical Because of Medical Necessity or Physical Availability (Table 1)*

Whether the pile drivers in Oak Ridge like it or not, half-hour $^{38}Cl$ is a practical radioisotope daughter. Chlorine-36 has such a long life that its specific activity, 31 $\mu g/\mu c$ (33 mc/g), puts it out of the true tracer category. The half-hour $^{34}Cl$ does not have a usable parent. A half-hour half-life for $^{38}Cl$ does not encourage anything but very quick studies. The 2.9-hr $^{38}S$ parent is even worse to contemplate. The element, however, is so essential in medical work that the production of $^{38}S$ cows of large curiage is not beyond reason.

Potassium-42 is not much better, but the greater than 3.5-year (recent data, about 35 years) $^{42}Ar$ parent increases its desirability. This fact added to the medical usefulness of potassium as a common electrolyte and its transient tumor uptake puts potassium in a class by itself.

Krypton is becoming more useful if only for the study of the movements of gases in respiratory passages. Because most pulmonary diagnostic tests need not involve long periods, the less than 2-hr half-life of $^{83m}Kr$ is not too great an obstacle.

The isotope $^{194}Au$ has almost the ideal characteristics for a radioisotope cow. In addition, the colloid is applicable to many circulation tests that attempt to circumvent metabolic behavior. Radiogold is adaptable to diagnostic tests involving any reticuloendothelial tissue, such as liver, spleen, or lymph nodes. Much of the therapeutic use of $^{198}Au$ has been preempted by chemicals easier to obtain and devoid of federal inspection, but some of the decline has been due to short shelf life. At the same time there is increasing evidence that much of the radiogold therapeutic usefulness was because of its rapid decay. Radioisotope cows that could produce $^{194}Au$ in multimillicurie amounts are practical.

The 6-hr half-life of the $^{99m}Tc$ daughter is reasonable for medical radioisotope scanning, but the 67-hr half-life of the $^{99}Mo$ parent is neither ideal nor reasonable for a cow. The isotope was selected for the low energy of its radiation. As most users know, low-energy radiation is easier to collimate. This isotope marks a historical end to the reign of physicists who think the human body should adapt itself to the rigors of the Chart of Nuclides. When J. G. Hamilton first started looking at radioisotopes a third of a century ago, he was interested in what the organism did with a chemical, not in how the body could be forced to fit some physical principles. Enough different radioisotopes of different elements are now available that the primacy of the body as a chemical organism

Table 1 RADIOISOTOPE COWS THAT ARE PRACTICAL

| No. | Mode of decay | Parent (P) Isotope $^{Z}_{A}$Symbol | Parent (P) Isotope Half-life | Parent (P) Shielding Radiation | Parent (P) Shielding Amount | Daughter (D) Isotope $^{Z}_{A}$Symbol | Daughter (D) Isotope Half-life | Predominant energy, Mev: $\beta^+$, % (approx.) | $\beta$ energy: High (>2) | $\beta$ energy: Medium (<2) | $\beta$ energy: Low (<1) | $\gamma$ energy: High (>1) | $\gamma$ energy: Medium (<1) | $\gamma$ energy: Low (<0.15) | Stable (S) $^{Z}_{A}$Symbol | Other isotope of daughter may be better? | Probable medical usefulness of daughter |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| **Because of Medical Necessity** | | | | | | | | | | | | | | | | | |
| 1 | P → D, D → S | $^{38}_{16}$S | 2.9 hr | $\beta^-$, $\gamma$ | Much | $^{38}_{17}$Cl | 38 min | | X | | | X | | | $^{38}_{18}$Ar | Very long $\beta^-$ and short $\beta^-$ and $\beta^+$ | Very |
| 2 | P → D → S | $^{42}_{18}$Ar | 3.5 years | $\beta^-$ ($\gamma$?) | Much | $^{42}_{19}$K | 12.5 hr | | X | | | X | | | $^{42}_{20}$Ca | Very long $\beta^-$ and short $\beta^-$ | Very |
| 3 | P → D, P → S | $^{83}_{37}$Rb | 83 days | $\epsilon$, $\gamma$ | Much | $^{83m}_{36}$Kr | 1.9 hr | | | | X | | | X | $^{83}_{36}$Kr | Yes | Very |
| 4 | P → D → S | $^{132}_{52}$Te | 3.2 days | $\beta^-$, $\gamma$ | Moderate | $^{132}_{53}$I | 2.3 hr | | | X | | | X | | $^{132}_{54}$Xe | Yes | Very |
| 5 | PP, PD, D → S | $^{194}_{80}$Hg | 130 days | $\epsilon$ | Moderate | $^{194}_{79}$Au | 1.7 days | 3 | | | X | | X | | $^{194}_{78}$Pt | Yes | Very |
| **Because of Physical Availability** | | | | | | | | | | | | | | | | | |
| 6 | P → D, GD, D → S | $^{90}_{38}$Sr | 28 years | $\beta^-$ | Little | $^{90}_{39}$Y | 2.7 days | | X | | | Very weak | | | $^{90}_{40}$Zr | Yes | Some |
| 7 | P → D (87%), P → S (13%), D → S | $^{99}_{42}$Mo | 2.8 days | $\beta^-$, $\gamma$ | Little | $^{99m}_{43}$Tc | 6 hr | | | | | | | | $\epsilon^{99}_{43}$Tc, $\epsilon^{99}_{41}$Ru | Yes | Little |
| 8 | P → D → S | $^{140}_{56}$Ba | 12.8 days | $\beta^-$, $\gamma$ | Much | $^{140}_{57}$La | 1.7 days | | | X | | X | | | $^{140}_{58}$Ce | Yes | Minor |

and the subsidiary nature of radioisotopes as a tool are beginning to be reasserted.

However, this does not imply that the molybdenum–technetium cow is useless. It is available; therefore it is a practical cow, at least physically practical (biological practicality remains to be seen). The same can be said of the strontium–yttrium and the barium–lanthanum cows.[8,9] These have been available for a long time. Each has characteristics adaptable to specific medical usefulness, especially where the metabolism of the element is of little concern. These are cows of great potentiality.

## Radioisotope Cows That May Be Practical Because of the Short Life of the Daughters (Table 2)

Eleven radioisotope cows have products with half-lives ranging from a few to many minutes. Their parents have half-lives ranging from as little as a day to $^{137}Cs$ at about 30 years.

Many things besides *clinical diagnosis* are involved in a *clinical radioisotope diagnosis*. The simplest, for example, is illustrated by the cesium–barium cow (No. 12 in Table 2). It can be used to demonstrate half-life. Those of you who know what the word *half-life* means do not need any further illustration; however, half-life is a complex subject, outside the realm of ordinary awareness. Once half-life has been demonstrated, it is easily understood and not forgotten by technicians. But the population of technicians has a half-life of 1 to 2 years, and so the demonstration must be repeated over and over.

Only a few of the very-short-lived cows have an immediate obvious metabolic applicability. The first minute or so of thyroid uptake of iodine is already a diagnostic test of some clinical significance. The same might easily be true of any element. This possibility gives the short-life (minute) cows an advantage in terms of iteration, redundancy (this is not an evil word), and multiple isotope duplication that could be decisive in some diagnostic tests.

The easy availability of multicurie parents means that these cows should be thought of for therapeutic as well as for diagnostic purposes. The daughters are ideal radioisotopes for some studies of radiobiological effects that cannot be accomplished with any other material. Some diagnostic tests (e.g., cardiac output) are done with the specific proviso that there be no mixing in the bloodstream and no metabolic activity.

## Radiosotope Cows That May Not Be Practical Because of the Short Life (Hours) of the Daughters (Table 3)

Everything that was said about the radioisotope cows that yield daughters with half-lives of minutes also applies to half-lives of hours, but there is a more likely metabolic interference with the longer half-life ones. The elements represented do not have a medical usefulness today, but this situation could change.[8,9] Also, with half-life in the hours range, there is time for some chemical manipulation before injection. Few of the moving-collimator scanning procedures are going to find such short-lived daughters useful, but all the stationary-collimator scanning procedures and many early single-shot measurement procedures (still the majority of diagnostic tests) are not affected by the half-life.

The old decisions concerning therapeutic use of $^{72}Ga$ are a good example of the difficulties in interpreting values. When used for therapy of bone tumors, $^{72}Ga$ has a large uptake in areas of new bone growth. However, the large uptake occurs after a few days have passed, and by this time the radioactive material has decayed; therefore the isotope is not useful for therapeutic purposes, but this judgment has nothing to do with the value of gallium for diagnostic purposes.

I would expect $^{68}Ga$ (No. 21, Table 3) to be useless* for diagnostic localization of bone tumors, and thus we lose the potential benefits of positron emission.[10] I would expect $^{72}Ga$ (No. 20, Table 3) to be useful under some conditions, and a $^{72}Ga$ cow might be important in areas where $^{67}Ga$ is not available. My statement that an element has little probable medi-

*R. Hayes of the Oak Ridge Institute of Nuclear Studies now seems to be disproving this statement (unpublished work, 1965).

Table 2 RADIOISOTOPE COWS THAT MAY BE PRACTICAL BECAUSE OF SHORT-LIFE (MINUTES) DAUGHTERS

| No. | Mode of decay | Parent (P) Isotope $^{Z}_{A}$Symbol | Parent (P) Isotope Half-life | Parent (P) Shielding Radiation | Parent (P) Shielding Amount | Daughter (D) Isotope $^{Z}_{A}$Symbol | Daughter (D) Isotope Half-life | $\beta^+$, % (approx.) | Predominant energy, Mev: $\beta$ energy High (>2) | $\beta$ energy Medium (<2) | $\beta$ energy Low (<1) | $\gamma$ energy High (>1) | $\gamma$ energy Medium (<1) | $\gamma$ energy Low (<0.15) | Stable (S) $^{Z}_{A}$Symbol | Other isotope of daughter may be better? | Probable medical usefulness of daughter |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 9 | P→D→S | $^{28}_{12}$Mg | 21 hr | $\beta^-$, $\gamma$ | Much | $^{28}_{13}$Al | 2.3 months | | X | | | X | | | $^{28}_{14}$Si | No | Little |
| 10 | P→D→S | $^{66}_{28}$Ni | 2.3 days | $\beta^-$ | Much | $^{66}_{29}$Cu | 5.2 months | | X | | | X | | | $^{66}_{30}$Zn | Yes | Some |
| 11 | P→D→S, P→GD→S | $^{122}_{54}$Xe | 20 hr | $\epsilon$, $\gamma$ | Much | $^{122}_{53}$I | 3.5 months | 100 | X | | | | X | | $^{122}_{52}$Te | Yes | Very |
| 12 | P→D→S, P→S | $^{137}_{55}$Cs | 30 years | $\beta^-$ | Moderate | $^{137m}_{56}$Ba | 2.6 months | | | | X | | X | | $^{137}_{56}$Ba | Yes | Little |
| 13 | P→D→S | $^{128}_{56}$Ba | 2.4 days | $\epsilon$, $\gamma$ | Much | $^{128}_{55}$Cs | 3.4 months | 75 | X | | | | X | | $^{128}_{54}$Xe | Yes | Little |
| 14 | P→D→S | $^{134}_{58}$Ce | 3 days | $\epsilon$ | Moderate | $^{134}_{57}$La | 6.5 months | 44 | X | | | | X | | $^{134}_{56}$Ba | Yes | Minor |
| 15 | P→D→S | $^{144}_{58}$Ce | 290 days | $\beta^-$, $\gamma$ | Much | $^{144}_{59}$Pr | 18 months | | X | | | ? | X | | $^{144}_{60}$Nd | Yes | Little |
| 16 | P→D→S | $^{140}_{60}$Nd | 3.3 days | $\epsilon$ | Little | $^{140}_{59}$Pr | 3.4 months | 58 | X | | | | X | | $^{140}_{58}$Ce | Yes | Little |
| 17 | P→D→S, P→GD→S | $^{160}_{68}$Er | 1.3 days | $\epsilon$ | Little | $^{160}_{67}$Ho | 25 months | < 1 | | | X | | X | | $^{160}_{66}$Dy | Yes | Little |
| 18 | P→D→S, P→GD→S | $^{178}_{74}$W | 22 days | $\epsilon$ | Little | $^{178m}_{73}$Ta | 9.4 months | < 2 | | | X | | | X | $^{178}_{72}$Hf | Yes | Little |
| 19 | P→D→S, P→S | $^{207}_{83}$Bi | 28 years | $\epsilon$, $\gamma$ | Much | $^{207m}_{82}$Pb | 1.4 months | | | | X | | X | | $^{207}_{82}$Pb | ? | Some |

Table 3 RADIOISOTOPE COWS THAT MAY NOT BE PRACTICAL BECAUSE OF SHORT-LIFE (HOURS) DAUGHTERS

| No. | Mode of decay | Parent (P) | | | | Daughter (D) | | | | | | | | | | Other isotope of daughter may be better? | Probable medical usefulness of daughter |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | Isotope | | Shielding | | Isotope | | | Predominant energy, Mev | | | | | | Stable (S) | | |
| | | | | | | | | | β energy | | | γ energy | | | | | |
| | | Symbol | Half-life | Radiation | Amount | Symbol | Half-life | β+, % (approx.) | High (> 2) | Medium (< 2) | Low (< 1) | High (> 1) | Medium (< 1) | Low (< 0.15) | Symbol | | |
| 20 | P →(1%) D; D → GD, S | $^{72}_{30}$Zn | 2 days | $\beta^-$ | Much | $^{72}_{31}$Ga | 14 hr | | | | X | X | | | $^{72}_{32}$Ge | Yes | Little |
| 21 | P → D → S | $^{68}_{32}$Ge | 275 days | $\epsilon$ | Much | $^{68}_{31}$Ga | 1 hr | 84 | | X | | X | | | $^{68}_{30}$Zn | Yes | Little |
| 22 | P →(1%) D, P → D; D → S | $^{82}_{38}$Sr | 26 days | $\epsilon$ | Much | $^{82m}_{37}$Rb | 6.3 hr | 21 | X | | X | | X | | $^{82}_{36}$Kr | Yes | Some |
| 23 | PP, PD → D; D → S | $^{87}_{39}$Y | 3.3 days | $\epsilon, \beta^+, \gamma$ | Much | $^{87m}_{38}$Sr | 2.8 hr | | | | X | | X | | $^{87}_{38}$Sr | Yes | Little |
| 24 | P → D; D → S | $^{103}_{44}$Ru | 41 days | $\beta^-, \gamma$ | Much | $^{103m}_{45}$Rh | 54 min | | | | X | | | X | $^{103}_{45}$Rh | Yes | Little |
| 25 | P →(99%) D; D → S | $^{103}_{46}$Pd | 17 days | $\epsilon, \gamma$ | Little | $^{103m}_{45}$Rh | 54 min | | | | X | | | X | $^{103}_{45}$Rh | Yes | Little |
| 26 | 1 P → D; 2 P → D; D →(5%) B, D → S | $^{115}_{48}$Cd | 2.2 days | $\beta^-, \gamma$ | Much | $^{115m}_{49}$In | 4.5 hr | | | | X | | X | | $^{115}_{49}$In, $^{115}_{50}$Sn | Yes | Little |
| 27 | PP, PD → D; D → S | $^{113}_{50}$Sn | 115 days | $\epsilon, \gamma$ | Moderate | $^{113m}_{49}$In | 1.8 hr | | | | X | | X | | $^{113}_{49}$In | Yes | Little |
| 28 | P → D; D →(4%) GD, D → GD; GD → S, GD → S | $^{141}_{62}$Sm | 20 days | $\beta^+$ | Much | $^{141}_{60}$Nd | 2.4 hr | 2 | | | X | X | | X | $^{141}_{59}$Pr | Yes | Little |
| | | | | | | $^{141}_{61}$Pm | 20 min | 100 | | X | | | X | | | | |
| 29 | P →(99%) D; D → GD, S | $^{166}_{70}$Yb | 2.4 days | $\epsilon$ | Moderate | $^{166}_{69}$Tm | 8 hr | <1 | | | X | | | X | $^{166}_{68}$Er | Yes | Little |
| 30 | P → D, S | $^{189}_{77}$Ir | 11 days | $\epsilon, \gamma$ | Moderate | $^{189m}_{76}$Os | 5.7 hr | | | | X | | | X | $^{189}_{76}$Os | Yes | Little |

Table 4 RADIOISOTOPE COWS THAT WOULD BE PRACTICAL

| No. | Mode of decay | Parent (P) Isotope Symbol | Half-life | Shielding Radiation | Amount | Daughter (D) Isotope Symbol | Half-life | $\beta^+$, % (approx.) | Predominant energy, Mev: $\beta$ energy High (>2) | Medium (<2) | Low (<1) | $\gamma$ energy High (>1) | Medium (<1) | Low (<0.15) | Stable (S) Symbol | Other isotope of daughter may be better? | Probable medical usefulness of daughter |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | | | **If Required for Medical Use** | | | | | | | | | | | |
| 31 | P→D(<2% GD), D→S | $^{72}_{34}$Se | 8.5 days | $\epsilon, \gamma$ | Moderate | $^{72}_{33}$As | 1 day | 30 | X | | | | X | | $^{72}_{32}$Ge | Yes | Some |
| 32 | P(<2% D), P→S, D→S | $^{95}_{40}$Zr | 63 days | $\beta^-, \gamma$ | Much | $^{95}_{41}$Nb | 35 days | | | | X | | X | | $^{95}_{42}$Mo | Yes | Little |
| 33 | P→D→S | $^{100}_{46}$Pd | 4 days | $\epsilon, \gamma$ | Much | $^{100}_{45}$Rh | 21 hr | 5 | | X | | | X | | $^{100}_{44}$Ru | Yes | Little |
| 34 | 1 P→D, 2 P→D, D→S (GD) | $^{119m}_{52}$Te | 4.6 days | $\epsilon, \gamma$ | Moderate | $^{119}_{51}$Sb | 1.6 days | | | | X | | | X | $^{119}_{50}$Sn | Yes | Some |
| 35 | P(83% D), P→S | $^{125}_{51}$Sb | 2.4 years | $\beta^-, \gamma$ | Much | $^{125m}_{52}$Te | 58 days | | | | X | | | X | $^{125}_{52}$Te | Yes | Little |
| 36 | P→D→S | $^{172}_{72}$Hf | 5 years | $\epsilon, \gamma$ | Moderate | $^{172}_{71}$Lu | 6.7 days | | | | X | | X | | $^{172}_{70}$Yb | Yes | Some |
| 37 | P→D, D→S (GD) | $^{188}_{78}$Pt | 10.3 days | $\epsilon, \gamma$ | Moderate | $^{188}_{77}$Ir | 1.7 days | <1 | | | X | | X | | $^{188}_{76}$Os | Yes | Little |
| | | | | | | **If Highly Desired** | | | | | | | | | | | |
| 38 | P→D→S | $^{112}_{46}$Pd | 21 hr | $\beta^-, \gamma$ | Moderate | $^{112}_{47}$Ag | 3 hr | | X | | | | X | | $^{112}_{48}$Cd | Yes | Some |
| 39 | PP, PD(<2% D), D→S | $^{111}_{47}$Ag | 7.5 days | $\beta^-, \gamma$ | Moderate | $^{111m}_{48}$Cd | 49 min | | | | X | | X | | $^{111}_{48}$Cd | Yes | Little |
| 40 | P(<2% D), P→S, D→S | $^{118}_{52}$Te | 6 days | $\epsilon$ | Little | $^{118}_{51}$Sb | 5 hr | Weak? | X | | | | X | | $^{118}_{50}$Sn | Yes | Some |
| 41 | P→D(25% GD), D→S | $^{200}_{78}$Pt | 11.5 hr | $\beta^-$ | Moderate | $^{200}_{79}$Au | 48 min | | X | | | | X | | $^{200}_{80}$Hg | Yes | Very |
| 42 | P(20% D), P→S | $^{204}_{83}$Bi | 12 hr | $\epsilon, \gamma$ | Moderate | $^{204m}_{82}$Pb | 1 hr | | | | X | | X | | $^{204}_{82}$Pb | ? | Some |
| | | | | | | **If Urgently Needed** | | | | | | | | | | | |
| 43 | P→D→S | $^{165}_{69}$Tm | 1.2 days | $\epsilon, \gamma$ | Little | $^{165}_{68}$Er | 10 hr | | | | X | | X | | $^{165}_{67}$Ho | Yes | Little |
| 44 | P→D, D→S, D→S (GD) | $^{188}_{74}$W | 65 days | $\beta^-$ | Moderate | $^{188}_{75}$Re | 17 hr | | X | | | | X | | $^{188}_{76}$Os | Yes | Little |
| 45 | P→D, D→S, D→S (GD) | $^{182}_{76}$Os | 1 day | $\epsilon$ | Moderate | $^{182m}_{75}$Re | 13 hr | | | | X | | X | | $^{182}_{74}$W | Yes | Little |
| 46 | P(<2% D), P→S, D→S | $^{194}_{76}$Os | 2 years | $\beta^-$ | Much | $^{194}_{77}$Ir | 19 hr | | X | | | | X | | $^{194}_{78}$Pt | Yes | Little |

Table 5 RADIOISOTOPE COWS THAT ARE IMPRACTICAL

| No. | Mode of decay | Parent (P) Isotope Symbol | Parent (P) Isotope Half-life | Parent (P) Shielding Radiation | Parent (P) Shielding Amount | Daughter (D) Isotope Symbol | Daughter (D) Isotope Half-life | $\beta^+$, % (approx.) | Predominant energy, Mev: $\beta$ energy High (>2) | $\beta$ energy Medium (<2) | $\beta$ energy Low (<1) | $\gamma$ energy High (>1) | $\gamma$ energy Medium (<1) | $\gamma$ energy Low (<0.15) | Stable (S) Symbol | Other isotope of daughter may be better? | Probable medical usefulness of daughter |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| **Because of Long Parent Life** | | | | | | | | | | | | | | | | | |
| 47 | P→D→S | $^{32}_{14}Si$ | $7 \times 10^2$ years | $\beta^-$ | Little | $^{32}_{15}P$ | 14.3 days | | | X | | | | | $^{32}_{16}S$ | Yes | Very |
| 48 | P→D, P→D→S | $^{44}_{22}Ti$ | $1 \times 10^3$ years | $\epsilon, \gamma$ | Much | $^{44}_{21}Sc$ | 4 hr | 94 | | X | | X | | | $^{44}_{20}Ca$ | Yes | Little |
| 49 | P→D, P→D (<1%)→GD, D→S | $^{60}_{26}Fe$ | $3 \times 10^5$ years | $\beta^-, \gamma$ | Much | $^{60}_{27}Co$ | 5.3 years | | | X | | X | | | $^{60}_{28}Ni$ | Yes | Very |
| 50 | P→D (50%), P→S | $^{93}_{40}Zr$ | $1 \times 10^6$ years | $\beta^-$ | Little | $^{93m}_{41}Nb$ | 10 years | | | | X | | | X | $^{93}_{41}Nb$ | Yes | Little |
| 51 | PP, PD→D→S | $^{202}_{82}Pb$ | $3 \times 10^5$ years | $\epsilon$ | Moderate | $^{202}_{81}Tl$ | 12 days | | | | X | | X | | $^{202}_{80}Hg$ | Yes | Little |
| **Because of Production Hardship** | | | | | | | | | | | | | | | | | |
| 52 | P→D (<0.1%), P→S | $^{111}_{49}In$ | 2.8 days | $\epsilon, \gamma$ | Moderate | $^{111m}_{48}Cd$ | 49 min | | | | X | | X | | $^{111}_{48}Cd$ | Yes | Little |
| 53 | P→D→s→S | $^{146}_{64}Gd$ | 52 days | $\epsilon, \gamma$ | Much | $^{146}_{63}Eu$ | 4.4 days | 10 | | X | | | X | | $^{146}_{62}Sm$ | Yes | Little |

cal usefulness is only an indication of my biochemical ignorance.

## Radiosotope Cows That Would Be Practical If Needed (Table 4)

Sixteen possible radioisotope cows seem to be impractical for one reason or another, but the impracticability is relative to the degree of medical need. Seven of these cows are not really impractical; there just doesn't seem to be much need for them. For example, a couple of other feasible radioisotopes of arsenic, providing either positron or negatron emission, are fairly easy to obtain by simpler methods. However, if there is a requirement for an arsenic cow, one is available.

Five of the cows seem to be somewhat impractical purely from the standpoint of half-life. The daughter does not decay sufficiently fast, the parent is not sufficiently long, or the production unnecessarily duplicates a more easily produced long-lived radioisotope of the same element. With the one exception of necessary iteration (and possible exception of a necessary built-in redundancy), almost all tests done with short-lived isotopes can also be done with long-lived isotopes. In many procedures the longer life is not only more economical but is preferable from a purely logistics standpoint. These five cows, although probably impractical, are available if a fairly high degree of desire for their specific characteristics should develop.

The last four cows shown in Table 4 seem to be even more impractical—from the physical manufacturing characteristics of the parent, from the short life of the parent, or from the unknown characteristics of the parent. They are not impossible to make, but there would have to be urgent requirements to overcome the undesirable features. With Nos. 43 to 46, as with Nos. 38 to 42, there are better radioisotopes.

## Radioisotope Cows That Are Impractical (Tables 5 and 6)

Seven radioisotope cows seem totally impractical. Phosphorus-32 can be made from a 700-year $^{32}Si$ parent. I cannot think of any condition that would make cow production preferable to present methods. The same is true of $^{60}Co$; it could be made from a $^{60}Fe$ parent. Ten-year $^{93m}Nb$ could be produced from a 1.1 million-year $^{93}Zr$ parent; I can't think of a single reason why these would be either necessary or desirable.

The only two radioisotope cows that I put under the heading of impractical for production reasons are $^{146}Gd$ and $^{111}In$. The $^{111}In$ is not even a good cow. Good reactor engineers can complain that many of the radioisotopes I say are practical are just as difficult to produce as $^{146}Gd$. It might be far easier to produce a gadolinium–europium cow than an argon–

Table 6 RADIOISOTOPE COWS THAT ARE TOTALLY (?) IMPRACTICAL

| No. | Parent | | Daughter | |
|---|---|---|---|---|
| | Because of Short Parent or Daughter Life | | | |
| 54 | $^{52}_{22}Ti$ | 12 min | $^{52}_{23}V$ | 3.8 min |
| 55 | $^{62}_{30}Zn$ | 9.3 hr | $^{62}_{29}Cu$ | 9.8 min |
| 56 | $^{83}_{35}Br$ | 2.3 hr | $^{83}_{36}Kr$ | 1.9 hr |
| 57 | $^{88}_{36}Kr$ | 2.8 hr | $^{88}_{37}Rb$ | 18 min |
| 58 | $^{80}_{38}Sr$ | 1.8 hr | $^{80}_{37}Rb$ | 34 sec |
| 59 | $^{97}_{40}Zr$ | 17 hr | $^{97m}_{41}Nb$ | 1 min |
| 60 | $^{101}_{42}Mo$ | 15 min | $^{101}_{43}Tc$ | 14 min |
| 61 | $^{102}_{42}Mo$ | 12 min | $^{102}_{43}Tc$ | 5 sec |
| 62 | $^{94}_{44}Ru$ | 57 min | $^{94}_{43}Tc$ | 53 min |
| 63 | $^{106}_{44}Ru$ | 1 year | $^{106}_{45}Rh$ | 30 sec |
| 64 | $^{108}_{44}Ru$ | 4 min | $^{108}_{45}Rh$ | 18 sec |
| 65 | $^{98}_{46}Pd$ | 18 min | $^{98}_{45}Rh$ | 9 min |
| 66 | $^{109}_{46}Pd$ | 14 hr | $^{109m}_{47}Ag$ | 39 sec |
| 67 | $^{118}_{48}Cd$ | 49 min | $^{118}_{49}In$ | 5 sec |
| 68 | $^{114}_{52}Te$ | 16 min | $^{114}_{51}Sb$ | 3 min |
| 69 | $^{116}_{52}Te$ | 3 hr | $^{116}_{51}Sb$ | 1 hr |
| 70 | $^{126}_{56}Ba$ | 1.6 hr | $^{126}_{55}Cs$ | 1.6 min |
| 71 | $^{142}_{62}Sm$ | 1.2 hr | $^{142}_{61}Pm$ | 34 sec |
| 72 | $^{158}_{68}Er$ | 2.4 hr | $^{158}_{67}Ho$ | 27 min |
| 73 | $^{161}_{68}Er$ | 3.5 hr | $^{161}_{67}Ho$ | 2.5 hr |
| 74 | $^{164}_{70}Yb$ | 1.2 hr | $^{164}_{69}Tm$ | 2 min |
| 75 | $^{190m}_{77}Ir$ | 3.2 hr | $^{190m}_{76}Os$ | 10 min |
| 76 | $^{192}_{80}Hg$ | 4.8 hr | $^{192}_{79}Au$ | 4.7 hr |
| | Because of Insufficient Parent-Daughter Time Differences* | | | |
| 77 | $^{47}_{20}Ca$ | 4.9 days | $^{47}_{21}Sc$ | 3.4 days |
| 78 | $^{86}_{40}Zr$ | 17 hr | $^{86}_{39}Y$ | 15 hr |
| 79 | $^{131}_{56}Ba$ | 12 days | $^{131}_{55}Cs$ | 10 days |
| 80 | $^{135}_{58}Ce$ | 22 hr | $^{135}_{57}La$ | 19 hr |

*See also No. 114, $^{172}_{68}Er$, in Table 9.

potassium cow, but $^{42}K$ is necessary and desirable, and no good substitutes are available. (Also, the cow is now being worked on.[7]) Europium has so many substitutes that the production problems can take precedence.

Table 6 shows 23 cows that have parents or daughters of such short half-life that the possibility of their use is doubtful. The $^{38}Cl$ (No. 1, Table 1) cow is practical because it is necessary; its only alternatives are radioisotopes that might be even more impractical to use. Copper-62 (No. 55, Table 6), $^{83}Kr$ (No. 56, Table 6), and the others are impractical for the same reason that $^{38}Cl$ is practical.

Four additional cows are impractical because there is so little difference between the half-lives of parent and daughter. It is possible to conceive of circumstances of pharmaceutical preparation or chemical composition that might make one of these practical, but not because of participation in a parent–daughter decay relation. (I am uncertain about the pharmacological uses of these four.)

### *Isomer Cows (Tables 7 and 8)*

True radioisotope cows (by the old definition) are not too unusual. With isomers, either the daughter cannot be separated from the parent, or the decay is from short- to long-lived. There is talk of a method of separating some isomer pairs,[7] but one is not yet available.

In some 130 instances (the exact number depends on how the words are defined), the daughters and parents are isomers. These might not be true radioisotope cows, but they follow the cow pattern and might be even more useful than ordinary radioisotopes. Five of these isomer cows decay from a long to a short half-life (Nos. 81 to 85, Table 7). Seven isomer cows decay from a short to a long half-life (Nos. 86 to 92, Table 7).

Six additional isomer cows undergo even further decay beyond the isomer, through a granddaughter stage (including Nos. 93 to 96, Table 8). There are 46 such possibilities, but most have some characteristic that makes them unsuitable for general clinical use (but not necessarily unsuitable for medical research).

### *Reverse Cows (Table 9)*

In 239 instances the relative times of parent and daughter decay are reversed. Most of these isotopes are impossible to use medically, but Table 9 shows 22 that have a reasonably useful life.

The potential importance of reverse cows in commercial production may be shown by an example. In the early days of the large-scale production of $^{131}I$, reactor production time was purchased for tellurium irradiation in a Canadian reactor. During shipment to American pharmaceutical houses, the 1.3-day tellurium parent decayed to the desired 8-day $^{131}I$ daughter, which was readily separated. Emphasis was on a pharmaceutically pure rather than a radioactively pure material.

But the usefulness of reverse cows is not limited to so simple a case. For example, production of a label that originates at the site of deposition of another label (with both physical and chemical localization) is an ideal pharmacological situation. These reverse cows

Table 7 ISOMER RADIOISOTOPE COWS

| No. | Mode of decay | Parent half-life to daughter half-life |
|---|---|---|
| | With Long to Short Life | |
| 81 | PP → S; PD → S | $^{44m}_{21}Sc$ 2.4 days → 4 hr |
| 82 | PP → S (90%); PD → S | $^{108m}_{47}Ag$ 5 years → 2 min |
| 83 | PP → S (3.5%); PD → S | $^{114m}_{49}In$ 49 days → 1.2 min |
| 84 | PP → S; PD → S | $^{121m}_{52}Te$ 154 days → 17 days |
| 85 | PP → S (2%); PD → S | $^{127m}_{52}Te$ 105 days → 9 hr |
| | With Short to Long Life | |
| 86 | PP → S; PD → S | $^{95m}_{41}Nb$ 3.5 days → 35 days |
| 87 | PP → S (?%); PD → S | $^{101m}_{45}Rh$ 5 days → 5 years |
| 88 | PP → S; PD → S | $^{133m}_{54}Xe$ 2.3 days → 5.3 days |
| 89 | PP → S; PD → S | $^{133m}_{56}Ba$ 1.6 days → 7.2 years |
| 90 | PP → S (?); PD → S | $^{184m}_{75}Re$ 2.2 days → 50 days |
| 91 | PP → S; PD → S | $^{193m}_{78}Pt$ 3.5 days → 500 years* |
| 92 | PP → D; PD → S | $^{197m}_{80}Hg$ 1 day → 2.7 days |

*This is probably not really a cow.

Table 8 ISOMER RADIOISOTOPE COWS WITH GRANDDAUGHTER CHARACTERISTICS*

| No. | Mode of decay | Parent Symbol | Parent Half-life | Daughter Symbol | Daughter Half-life | Stable | Remarks |
|---|---|---|---|---|---|---|---|
| 93 | PP → PD (D); GD, S | $^{129m}_{52}$Te | 33 days | $^{129}_{53}$I | $1.7 \times 10^7$ years | $^{129}_{54}$Xe | Might be useful in chronic injection experiments |
| | | $^{129}_{52}$Te | 74 min | | | | |
| 94 | PP → PD (D); GD, S, GD | $^{131m}_{52}$Te | 1.3 days | $^{131m}_{54}$Xe | 12 days | $^{131}_{54}$Xe | Production method for $^{131}_{53}$I |
| | | $^{131}_{52}$Te | 25 min | $^{131}_{53}$I | 8 days | | |
| | | | | $^{131m}_{54}$Xe | Very short | | |
| 95 | PP → PD (D → S) | $^{137m}_{58}$Ce | 1.5 days | $^{137}_{57}$La | $6 \times 10^4$ years | $^{137}_{56}$Ba | Probably too short-lived PD for anything but specific research use |
| | | $^{137}_{58}$Ce | 9 hr | | | | |
| 96 | PD → D; PD → D → S | $^{195m}_{80}$Hg | 1.7 days | $^{195m}_{79}$Au | 30 sec | $^{195}_{78}$Pt | Production method for $^{195}_{79}$Au |
| | | $^{195}_{80}$Hg | 10 hr | $^{195}_{79}$Au | 185 days | | |

*There are 46 possibilities with half-lives too short to consider; $^{119m}_{52}$Te is No. 34 in Table 4, $^{99}_{42}$Mo is No. 7 in Table 1; No. 94 in this table is also No. 105 in Table 9.

Table 9 REVERSE RADIOISOTOPE COWS THAT MAY BE PRACTICAL

| No. | Parent | | Daughter | |
|---|---|---|---|---|
| 97 | $^{48}_{24}$Cr | 1 day | $^{48}_{23}$V | 16 days |
| 98 | $^{55}_{27}$Co | 18 hr | $^{55}_{26}$Fe | 2.9 years |
| 99 | $^{56}_{28}$Ni | 6.4 days | $^{56}_{27}$Co | 77 days |
| 100 | $^{57}_{28}$Ni | 1.5 days | $^{57}_{27}$Co | 270 days |
| 101 | $^{83}_{38}$Sr | 1.5 days | $^{83}_{37}$Rb | 83 days |
| | | | $^{83m}_{36}$Kr(GD) | 1.9 hr |
| 102 | $^{88}_{40}$Zr | 85 days | $^{88}_{39}$Y | 105 days |
| 103 | $^{125}_{50}$Sr | 9.5 days | $^{125}_{51}$Sb | 2.4 years |
| | | | $^{125m}_{52}$Te(GD) | 58 days |
| 104 | $^{127}_{51}$Sb | 3.9 days | $^{127m}_{52}$Te | 105 days (complex) |
| 105 | $^{131}_{52}$Te | 1.3 days | $^{131}_{53}$I | 8 days (complex) |
| 106 | $^{143}_{58}$Ce | 1.4 days | $^{143}_{59}$Pr | 14 days |
| 107 | $^{147}_{60}$Nd | 11 days | $^{147}_{61}$Pm | 2.5 years |
| 108 | $^{151}_{61}$Pm | 1.1 days | $^{151}_{62}$Sm | 100 years |
| 109 | $^{145}_{63}$Eu | 5.6 days | $^{145}_{62}$Sm | 1 year |
| | | | $^{145}_{61}$Pm(GD) | 18 years |
| 110 | $^{147}_{64}$Gd | 1.2 days | $^{147}_{63}$Eu | 24 days (complex) |
| 111 | $^{149}_{64}$Gd | 9 days | $^{149}_{63}$Eu | 120 days |
| 112 | $^{153}_{65}$Tb | 2.6 days | $^{153}_{64}$Gd | 236 days |
| 113 | $^{166}_{66}$Dy | 3.3 days | $^{166}_{67}$Ho | 30 years (complex) |
| 114* | $^{172}_{68}$Er | 2.1 days | $^{172}_{69}$Tm | 2.6 days |
| 115 | $^{169}_{71}$Lu | 1.7 days | $^{169}_{70}$Yb | 32 days |
| 116 | $^{173}_{72}$Hf | 1 day | $^{173}_{71}$Lu | 200 days |
| 117 | $^{181}_{75}$Re | 20 hr | $^{181}_{74}$W | 140 days |
| 118 | $^{195m}_{80}$Hg | 1.7 days | $^{195}_{79}$Au | 185 days (complex) |

*See also Table 6.

and especially the reverse isomer cows may well turn out to be as important in pharmacology as the original discovery of the isotope label.

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Section I

Isotopes and Radiation Technology

# Isotope Production and Development

## The Western New York Nuclear Research Center

By C. C. Thomas, Jr., D. A. Berry, P. M. Orlosky, and G. P. Tercho

*The Western New York Nuclear Research Center, State University of New York, produces short-lived isotopes, provides service irradiations, and carries out research in radiation chemistry, activation analysis, and radiation effects. The facilities, which are described, include a reactor capable of pulsed operation and two Van de Graaff accelerators.*

The Western New York Nuclear Research Center, Inc., is a wholly owned subsidiary of the State University of New York and is located on the Buffalo campus of the University. The essential purpose of the Center is to provide the highly specialized tools and services necessary to support the nuclear education and research programs of the educational, industrial, medical, and governmental communities. To accomplish its objectives, the Center is equipped with a nuclear research reactor, PULSTAR, which has a pulsing capability, and two Van de Graaff accelerators. The Center has the laboratories, hot cell, and counting facilities necessary to support nuclear research and development. A full-time professional staff operates and maintains the facilities. This includes personnel for health physics, for electronics repair, and for a machine shop.

### Service and Research Activities

The activities of the Center cover service irradiations, isotope production, and research in radiation chemistry, activation analysis, and radiation effects.

The facilities of the Center are available to qualified research and developmental groups. Staff personnel assist individuals and companies using the Center's facilities for service irradiations. This assistance ranges from minimal aid, such as dosimetry and sample insertions, to complete responsibility for an entire experimental program.

Isotope production is primarily geared to short-lived isotopes because of the reactor operating schedule. However, longer lived isotopes can be obtained on request. As a general rule isotopes are produced on a custom basis to the users' specifications. Isotopes frequently requested are listed in Table I-1. Many of the more than 25 radioisotope laboratories on the campus depend partly upon short-lived isotopes generated at the Center.

Table I-1 ISOTOPES PREPARED AT THE WESTERN NEW YORK NUCLEAR RESEARCH CENTER, INC.

| Isotope | Half-life | Usual chemical form | Specific activity, μc/mg |
|---|---|---|---|
| $^{82}Br$ | 35.5 hr | KBr in HCl | 37 |
| $^{115}Cd$ | 53.5 hr | $Cd(NO_3)_2$ in $HNO_3$ | 40 |
| $^{64}Cu$ | 12.9 hr | $Cu(NO_3)_2$ in $HNO_3$ | 27 |
| $^{32}P$ | 14.3 days | $H_3PO_4$ in HCl | 16 or carrier-free |
| $^{42}K$ | 12.5 hr | KCl in HCl | 110 |
| $^{24}Na$ | 15.0 hr | NaCl in HCl | 1 |
| $^{86}Rb$ | 18.7 days | RbCl in HCl | 130 |
| $^{77}Ge$ | 12 hr | $GeCl_2$ in HCl | 19 |
| $^{124}Sb$ | 60.9 days | $SbCl_3$ in HCl | 7 |
| $^{18}F$ | 1.9 hr | NaF in NaCl solution | Carrier-free |
| $^{199}Au$ | 3.1 days | $AuCl_3$ in HCl + $HNO_3$ | Carrier-free |
| $^{90}Y$ | 64.4 hr | $Y_2O_3$ | 100 |

A number of the radioisotopes produced at the Center, such as $^{82}Br$, $^{18}F$ (Ref. 1), and $^{90}Y$ are used by outside medical organizations in their research, diagnostic, and therapeutic programs. Important applications for these isotopes include diagnosis of specific types of cancerous tissue and cancer therapy.

The Center also furnishes a number of other services, including activation analysis, envi-

ronmental surveys, bioassays, health physics instrument calibration, designs and surveys for radiation protection, and X-ray calibration. One area that is showing rapid growth is activation analysis. Because it is a powerful and sensitive technique for trace-element determination, activation analysis is used extensively by many industries for the analysis of a wide variety of sample materials. The application of many materials frequently depends on the very small quantities of impurities present in these materials. The technique of activation analysis not only allows identification of these trace impurities but also permits determination of the exact amounts present. Samples analyzed routinely include biological specimens, soils, electronic components, photographic film, refractory fiber materials, and alloys.

The Research Department carries out programs for industrial and government groups under contracts or grants. These projects may be handled wholly by staff personnel or may involve University faculty and graduate assistants. The range of activities is wide and includes work in tracer technology, activation analysis, and radiation chemistry. Projects now in progress include activation analysis of blood, a tracer study of corrosion in heat exchangers, the production of wood–plastic combinations with fission-product gamma sources, and a study related to the commercialization of wood–plastic combinations. The last project is supported in part by the Division of Isotopes Development, U. S. Atomic Energy Commission. A project to be started in the near future will deal with investigation of the usefulness in activation analysis of short-lived isotopes produced under pulsed reactor operation.[2]

In addition, the facilities of the Center extensively support the educational and research programs of the State University of New York, principally those of the School of Engineering and the Departments of Biology, Chemistry, and Physics. Research projects in fission-product chemistry,[3] neutron diffraction, nuclear decay-scheme studies, radiation biology, and activation analysis are in progress.

## Reactor Facilities

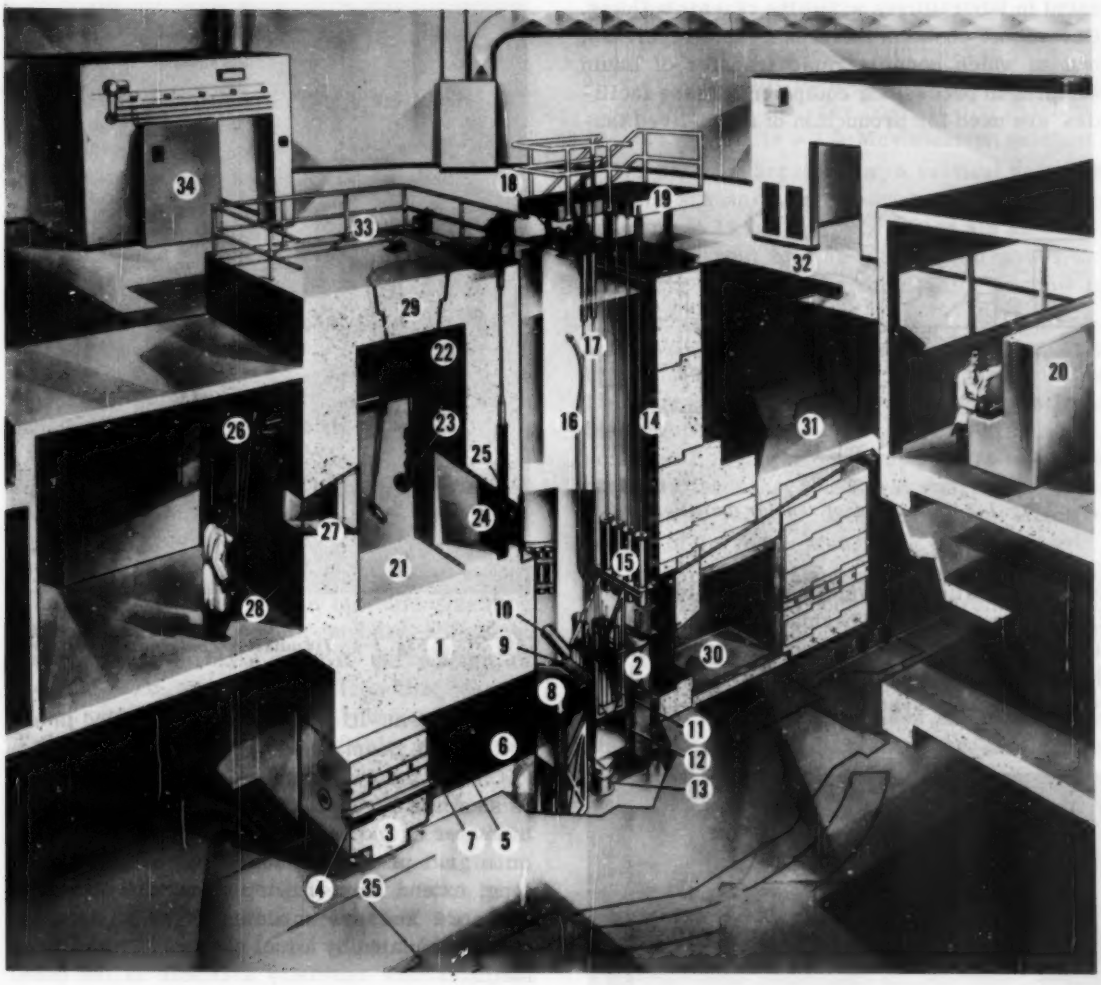
The reactor, PULSTAR, which started operation in June 1964, is a water-cooled and -moderated research reactor designed by AMF Atomics, Inc. It uses slightly enriched (6%) uranium in the form of sintered $UO_2$ pellets clad in Zircaloy tubes.[4] Twenty-five such tubes make up each fuel assembly, and nominally 20 assemblies constitute an operating core of roughly 15 by 15 by 24 in. The reactor, designed for 5-Mw operation, is operated presently at a steady-state power level of 2 Mw and can be pulsed at 10,000 Mw. The power level can be increased to 5 Mw with a slight modification of the cooling system. The program to investigate the pulsing characteristics of the reactor is still in progress; hence the ultimate pulse capability is not yet known. Sufficient data have been accumulated to justify application for a license to pulse routinely at 2000 Mw. The reactor characteristics are as follows:

| | |
|---|---|
| Operating power | |
| Steady state | 2 Mw |
| Pulse mode, initial | 2000 Mw (35 Mw-sec) |
| Pulse mode, design | 10,000 Mw (73 Mw-sec) |
| Thermal-neutron flux | |
| Steady state, in-core facilities | $4 \times 10^{13}$ n/(cm$^2$)(sec) maximum |
| Pulse mode, design, in-core facilities | $2 \times 10^{17}$ n/(cm$^2$)(sec) |
| Fast-neutron flux | |
| In-core facilities | $5 \times 10^{13}$ n/(cm$^2$)(sec) |
| Operating schedule | 16 hr/day, 5 days/week in normal operation; 3-shift operation when necessary |
| Operating time | About 80% |

The neutron irradiation facilities available for use are shown in Figs. I-1 and I-2. These facilities include beam tubes, pneumatic rabbits, isotope-production facilities, a thermal column, and a large dry irradiation chamber. Figure I-1 is an isometric drawing of the reactor core and experimental facilities. The reactor core, thermal column, and dry irradiation chamber are indicated at number positions 11, 6, and 30, respectively. Figure I-2 shows the facilities in plan view. Gamma irradiations are carried out in the hot cell (No. 21 in Fig. I-1), utilizing the fission-product activity of the fuel elements,[5] or in the large dry chamber, using residual gamma radiation after shutdown.

There are six 6-in. beam tubes and one 12-in.-square beam tube that can be used dry or water flooded. At 2 Mw, thermal-neutron fluxes of about $10^{13}$ n/(cm$^2$)(sec) with equivalent fluxes above 10 kev are available at the core end of these tubes.

Two pneumatic rabbit tubes presently used can handle $1\frac{7}{8}$-in.-diameter by $7\frac{1}{2}$-in.-long containers. These conveyors have a transit time of about 3 sec and terminate in fume hoods lo-




1. Biological Shield
2. Floodable Nosepiece
3. Thermal-Column Door
4. Shielding Plug
5. Boral Lining
6. Graphite Blocks (removable)
7. Experimental Space
8. Graphite Nosepiece
9. Lead Shield
10. Beam Tube
11. Reactor Core
12. Plenum
13. Coolant Discharge
14. Reactor-Tank Wall
15. Reactor-Control Instrumentation
16. Pneumatic Carrier Tube
17. Control-Rod Drives
18. Control-Rod-Drive Motors
19. Reactor Bridge
20. Reactor-Control Console
21. Hot Cell
22. Crane
23. Pass-Through Tube
24. Gamma Facility
25. Lead Shutter
26. Remote Manipulator
27. High-Density Viewing Window
28. Access Port
29. Removable Shielding Plug
30. Dry Exposure Chamber
31. Gamma Deck
32. Control Deck
33. Pass-Through Tube Controls
34. Air-Lock Door
35. Neutron Deck

*Fig. I-1 Isometric drawing of reactor facilities at Western New York Nuclear Research Center.*

cated in laboratories within the reactor building. One rabbit contains interchangeable receivers, one of which permits rapid transfer of liquid samples to processing equipment. These facilities are used for production of short-lived isotopes, activation analysis, and short-term controlled irradiations. Thermal-neutron fluxes of up to $5 \times 10^{12}$ n/(cm$^2$)(sec) are available at 2 Mw in either rabbit tube. A fast-transit-time (< 1 sec) rabbit system containing three independent $\frac{3}{4}$-in.-ID tubes is being installed in one of the 6-in. beam tubes.

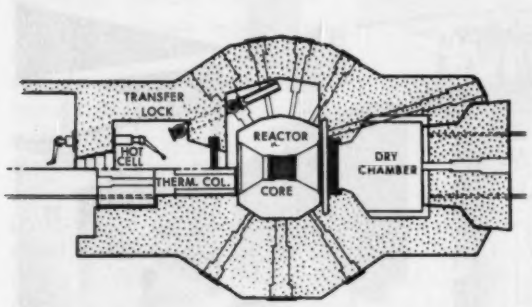



*Fig. I-2 Plan view of reactor facilities.*

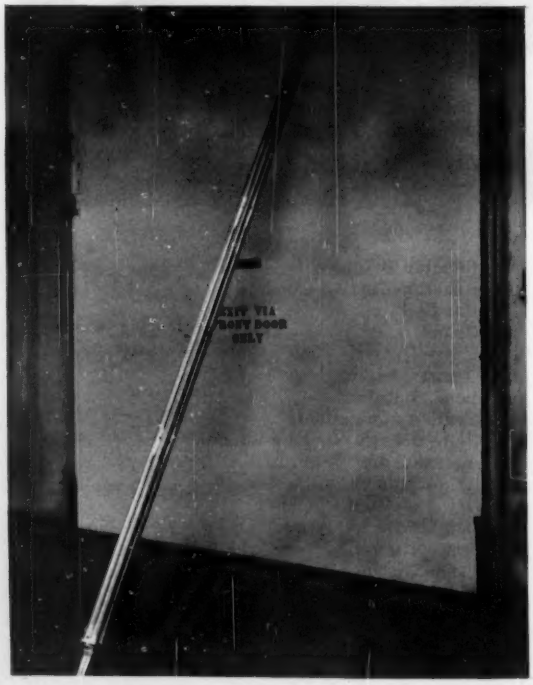


*Fig. I-3 Cylindrical tubes used as isotope-irradiation facility.*

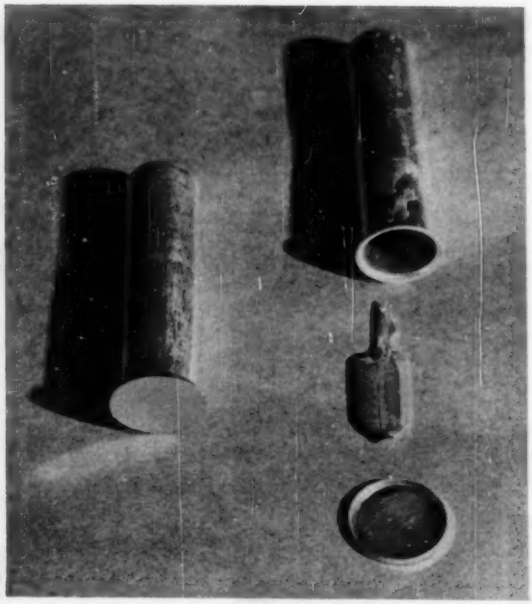


*Fig. I-4 Irradiation containers.*

One of the two isotope-irradiation facilities is shown in Fig. I-3. It consists of four $1\frac{1}{4}$-in.-diameter cylindrical tubes in a cluster, mounted on a grid plug. The tubes, approximately 25 ft long, extend from the top of the tank down into the core and are mounted so as to occupy a position vacated by a fuel element. Two of these facilities are normally available in the core. Typical containers are shown in Fig. I-4. The quartz primary containers allow relatively contamination-free encapsulation for most of the isotopes. However, primary containers of polyethylene are also used for some special applications. These containers are placed inside aluminum cans used for secondary containment, and the cans are sealed with a crimping tool. The thermal-neutron flux within the isotope facilities varies with core geometry and position and from tube to tube. The maximum flux available at 2 Mw is about $4 \times 10^{13}$ n/(cm$^2$)(sec).

The large dry exposure chamber (Fig. I-1, No. 30) has a $3\frac{1}{2}$-ft-square window just inside the tank wall, and just behind this space the chamber widens to 7 by 7 by 7 ft. Shielding is provided by a 6-ft-thick steel-encased high-density concrete door. The floodable nosepiece (No. 2, Fig. I-1), as well as a movable 4-ft by

4-ft by 10-in. lead shutter, provides shielding and allows entrance to the chamber when the reactor is not operating. Thermal-neutron fluxes up to $2 \times 10^{11}$ n/(cm$^2$)(sec) at 2 Mw are available in the exposure chamber. With the nosepiece flooded, relatively neutron-free gamma irradiations can be carried out in the chamber with the reactor operating. With the nosepiece voided, residual gamma radiation after shutdown can be used for gamma irradiations.

The graphite thermal column is a source of thermal neutrons free of fast neutrons. Immediately adjacent to the reactor core is a 6-in.-thick lead gamma shield. Between this shield and the reactor tank wall is an aluminum-clad graphite nosepiece, 2.5 ft thick. Beyond the tank wall, in place of the usual high-density concrete shield, is a cavity 4 by 4 by 6½ ft, stacked with graphite stringers. The working chamber is 4 by 4 by 1½ ft, but this can be expanded by withdrawing the movable shielding door 4 ft and adding some peripheral shielding. Stringers are broken so that the amount of graphite, and hence the neutron flux, can be varied quite easily. Thermal fluxes of about $.1 \times 10^9$ n/(cm$^2$)(sec) are available with a cadmium ratio of about $10^3$.

A fission plate, consisting of an aluminum–uranium (fully enriched) alloy containing about 1 kg of $^{235}$U and clad with 0.025-in. aluminum, is available for use in the thermal column or in the vestibule of the 12-in. beam tube. The plate is 13 in. square and ½ in. thick. This plate is used to provide relatively pure fission-spectrum neutrons.

The hot cell is connected to the reactor tank by a watertight pass-through lock. The lock permits transfer of samples up to about 5½ in. in diameter and 40 in. long from the core to the hot cell. Before the hot-cell end of the lock is opened for sample removal, any water introduced with the sample can be removed through the valving system. The cell capacity is the equivalent of 25 kilocuries of $^{60}$Co.

A second feature of the hot-cell design is a gamma facility that permits gamma irradiations using fuel elements. The gamma facility is a 2- by 2-ft cavity in the concrete shield between the cell and the reactor tank. Instantaneous gamma dose rates up to $8 \times 10^6$ rads/hr can be obtained by using fuel elements removed from the core immediately after shutdown from 2-Mw operation.[5]

## Accelerator Facilities

The Center is equipped with two Van de Graaff accelerators built by High Voltage Engineering Corp. One is a 400-kev positive-ion machine, and the second is a 1.5-Mev electron machine.

The electron accelerator, a vertical unit with a beam scanner that can produce a ⅜-in.-wide beam up to 15 in. long, is rated at 1.5 Mev with a beam current of 1.75 ma. At full power the radiation output at the accelerator window is about $2.5 \times 10^8$ rads/sec.

A gold target can be installed on the machine to produce an X-ray beam. At full power with a 15-in. scan, the radiation intensity 5 cm from the target is about $6 \times 10^5$ rads/hr. With a 6-in. scan the full-power output at 5 cm is in excess of $1 \times 10^6$ rads/hr. The X-ray energy spectrum has a peak value of 0.48 Mev and an effective energy of 0.8 to 0.9 Mev.

An overhead rail-and-chain conveyor system transports material continuously from a control room to a shielded target area. A maximum of 62 carriers can be attached to the conveyor. Each carrier has an area of about 2000 cm$^2$ (310 sq in.) and can handle a 60-lb load. The distance from the carrier to the accelerator window is adjustable from 4 to 69 cm. The conveyor speed can be varied from 2.7 to 100 cm/sec, and the conveyor direction is reversible. Limit switches can be set to oscillate automatically a small number of carriers back and forth under the beam. A floor-mounted truck and dolly, powered by the conveyor, can oscillate large experiments under the beam. Rotating sample holders and other special apparatus are also available. A remotely controlled shutter can be used to control short electron exposures precisely.

The positive-ion accelerator is primarily used as a neutron generator in a steady state or in a pulse mode. Maximum beam current is 150 μa. Two neutron-producing targets are used—a beryllium target to produce polyenergetic neutrons and a tritiated material to yield 14.7-Mev neutrons when bombarded with deuterons. The maximum total output of 14.7-Mev neutrons is about $10^{10}$ n/sec.

The drift tube of the accelerator extends through a removable shield into a small cave. The drift tube can can be removed from the cave without removing the shielding. A water-filled moderator tank is installed in the cave when desired. When the tank is used, the ac-

celerator drift tube leads into a compartmented beam tube centered in the tank. One section of the beam tube may be used either dry or wet when the rest of the tank is flooded. The sample tube of a small pneumatic rabbit system is located in this section of the beam tube and is separated from the target of the accelerator by a thin aluminum barrier. The pneumatic rabbit terminates in a detector housing in a counting room.

### Acknowledgments

The authors wish to thank William F. Hall, Dr. R. F. Lumb, James C. Evans, and Dr. C. H. Collins for their invaluable assistance in the preparation of this article.

# Computer Codes for Isotopes Calculations

By Charles W. Friend,* Alton R. Jenkins,* R. E. Lewis,* and J. R. Knight†

*Some of the basic characteristics of computer programs developed in the Isotopes Division at Oak Ridge National Laboratory are reviewed, and present descriptions of ISOTOPES, ISOCRUNCH, ISOPOWER, and ISOSEARCH are given.*

## Introduction

In isotopes production, calculations of such values as reactor yields and power densities of radioisotopic compounds are not mathematically complex but are rather tedious to perform. With the availability and increased flexibility of digital computer systems during the last decade, many calculations that would take months by hand can be performed in minutes on a modern computer. The processing of problems has become as routine and rapid as getting a shirt washed at a modern laundry; i.e., a problem submitted in the morning to a computing center can be processed and out by the afternoon.

Computer programs can be written to evaluate almost any equation or to perform any set of numerical operations that a man can do if he has the time. The computer simply relieves qualified personnel from performing certain time-consuming calculations over and over again. The flexibility of modern computer-program languages, e.g., FORTRAN, has made possible the writing of programs that can be run after minor adaptation on any modern computer. This means that, once a program which meets a basic need has been written, it can be used by other people at other installations with little change. It is possible at most computing centers to get the help of a trained programmer in planning and writing a program that will not only meet one's present needs but can also be changed as required. Different types of computer outputs are available, including printouts and graphical plots.

## Development of Computer Programs at ORNL

One of the basic considerations in the development of a code in the Oak Ridge National Laboratory (ORNL) Isotopes Division is the design of an input-data format that is easy to use and an output that is easy to read. The input data for a calculation are listed by the user in tabular form on a computer data sheet. The printed output is designed to reproduce this table along with the calculated results clearly labeled and listed in a manner that allows rapid interpretation. Graphical plots of the results are often available from associated equipment such as the CALCOMP or Benson–Lehrner electroplotters. Since all the codes have been written in FORTRAN language, problems may be processed at either the ORNL computing facility with the CDC 1604-A (Fig. I-5) or the Oak Ridge Gaseous Diffusion Plant Central

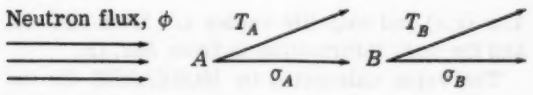
*Fig. I-5 Oak Ridge National Laboratory CDC 1604-A computing facility.*

*Oak Ridge National Laboratory, Oak Ridge, Tenn.
†Oak Ridge Gaseous Diffusion Plant, Oak Ridge, Tenn.

Data Processing Facility with the IBM 7090. In this review we present four of the programs that have been developed here for isotopes calculations. The authors would be most grateful if persons having information about other codes in this field (reports, abstracts, etc.) would send it to the ORNL Isotopes Information Center. It is hoped that a compilation can be made that could be published at a later date. Anyone who is interested in using one or more of these programs may write the authors for a copy of the FORTRAN deck and a report describing the program in detail.

*ISOTOPES*

The first program discussed is ISOTOPES,[6] which was developed for simple neutron-capture and decay problems. In a simple neutron-capture reaction, an isotope, $A$, having a neutron-capture cross section $\sigma_A$ may capture a neutron to form isotope $B$ of the same element. The isotope $B$ is usually radioactive with a half-life $T_B$ and may in turn have a significant neutron-capture cross section, $\sigma_B$. If isotope $A$ is radioactive, its half-life is designated $T_A$. A reaction and decay diagram for a general case is




Program ISOTOPES was developed to calculate the initial specific activity, in curies per gram, of the product isotope $B$ per gram of isotope $A$, following any irradiation period in the neutron flux, $\phi$, of a reactor. The program can also be used to find the optimum time of irradiation for maximum yield at any flux.

This program was written in FORTRAN 63 computer language for the CDC 1604-A computer. The input data are written in the form of a table, which is printed in the output along with the calculated results (Fig. I-6). Program ISOTOPES was used to calculate yields of 50 isotopes for three different flux values and various reactor irradiation times for a compilation in the ORNL Radioisotopes Procedures Manual.[7]

*ISOCRUNCH*

Program ISOCRUNCH[8] is a modification and expansion of a basic computer code CRUNCH.[9] The original CRUNCH code was used to calculate the amount of each isotope generated in a single reaction or decay chain of nuclides: $A$, $B$, $C$, ... up to 50. The program used a generalized solution of the Bateman equations,[10] which describe such processes. In developing ISOCRUNCH, the means of writing the input data were simplified and the printed output was made easier to read. An optional program was added to the code to find the total amount of any single nuclide produced by various contributing reactions and decay chains. With this program the optimum time of neutron irradiation for maximum yield of any particular nuclide can be found. A graphical plotting option is available to generate yield–time curves on a computer-associated electroplotter. ISOCRUNCH was originally written in FORTRAN II for the IBM 7090 with an associated Benson–Lehrner electroplotter and has been adapted to the CDC 1604-A computer with an associated CALCOMP plotter. A graphical plot of the yield of $^{60}$Co during a 10-year irradiation of $^{59}$Co at a neutron flux of $2 \times 10^{14}$ n/(cm$^2$)(sec) is shown in Fig. I-7.

*ISOPOWER*

ISOPOWER[11] is used to compute the maximum theoretical thermal power generated from the decay of a radioisotope as a function of time. The thermal output can be converted to electric power with a thermoelectric conversion system. Radioisotopes are now being used for power in ocean, arctic, and space devices, where an independent source of power is needed in a

| ISOTOPE | CROSS SECTION (BARNS) | HALF-LIFE | NP | I | WEIGHT (GRAMS) | FLUX | TMAX (DAYS) | PRODUCT ACT (C/G) | COMBINED ACT (C/G) |
|---|---|---|---|---|---|---|---|---|---|
| SB 121 | 6.00000E+00 | -0 | -0 | .57250 | 121.750 | 1.7000E+13 | 4.1339E+01 | 7.8804E+00 | 7.8804E+00 |
| SB 122 | 0 | 2.80000E+00D | | | | 3.2000E+14 | 2.9501E+01 | 1.4766E+02 | 1.4766E+02 |
| | | | | | | 2.0000E+15 | 2.2172E+01 | 9.8617E+02 | 9.8617E+02 |
| SB 121 * | 6.00000E+00 | -0 | -0 | .57250 | 121.750 | 1.7000E+13 | 7.0000E+00 | 6.4895E+00 | 6.4895E+00 |
| SB 122 * | 0 | 2.80000E+00D | | | | 3.2000E+14 | 7.0000E+00 | 1.2207E+02 | 1.2207E+02 |
| | | | | | | 2.0000E+15 | 7.0000E+00 | 7.5994E+02 | 7.5994E+02 |

*Fig. I-6 Program ISOTOPES printout for yield calculations at the optimum time (Tmax.) and at an arbitrary time for three flux values. All numbers are expressed as $1.7000_{10} + 13 = 1.7 \times 10^{13}$.*

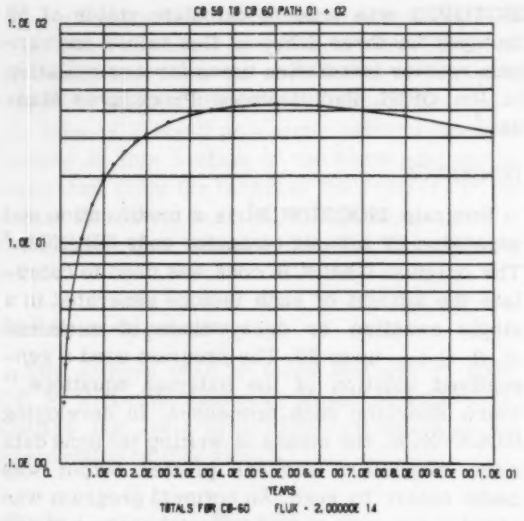


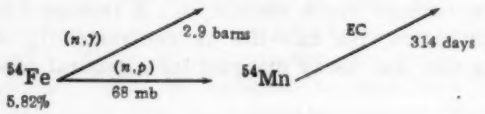

*Fig. I-7 Program ISOCRUNCH graph produced by Benson–Lehrner electroplotter for yield (at. %) of $^{60}Co$ during a 10-year irradiation of $^{59}Co$ at a neutron flux of $2 \times 10^{14}$ n/(cm²)(sec).*

remote region. The thermal power output of various radioisotope compounds will need to be calculated as these compounds are developed. If more than one radionuclide is present in a power-source compound, this program will sum all the contributions by the various nuclides to the total thermal output. A printout of results for a $^{90}Sr$ power-source compound is shown in Fig. I-8. If desired, power output–time curves can be plotted on a computer-associated electroplotter. This program has been programmed for the IBM 7090 in FORTRAN II and will be adapted to the CDC 1604-A in FORTRAN 63 language.

*ISOSEARCH*

Program ISOSEARCH is being developed to find an unknown reaction cross section, half-life, flux value, or product activity in a chain consisting of two or three nuclides. The searching method starts from maximum and minimum estimates, which are supplied, and finds a value for the unknown that fits the rest of the input data.

An example of its use would be to find the fast-neutron flux for a particular reactor position when all other information is known for the monitor used. Suppose an iron-wire monitor weighing 10 mg had been irradiated in a reactor for 30 days and the total activity for the $^{54}Mn$ formed had been determined to be $6.01 \times 10^6$ dis/sec. The reaction and decay scheme is




The $(n,\gamma)$ and half-life values are from Ref. 12, and the $(n,p)$ information is from Ref. 13.

The value calculated by ISOSEARCH for the fast-neutron flux for the $(n,p)$ reaction (flux above the reaction threshold energy) would be $2.2 \times 10^{14}$ n/(cm²)(sec).

ISOPOWER SR-90 ISOTOPIC POWER SOURCE

CHAIN NO. 1

| ISOTOPE | HALF LIFE | OP | I | WEIGHT | DENSITY | ENERGY (MEV) | AV. ENERGY (MEV) | TIMES | W/GM | POWER W/CC | CURIES/GM |
|---|---|---|---|---|---|---|---|---|---|---|---|
| SR 90 | 2.8000E 01Y | -1 | 0.2500 | 102.0000 | 4.7000 | 0.5400 | 0.2000 | 1.4000E 01D | 3.7076E-02 | 1.7426E-01 | 3.1277E 01 |
| | | | | | | | | 5.0000E 00Y | 3.2790E-02 | 1.5412E-01 | 2.7662E 01 |
| Y 90 | 6.4200E 01H | -1 | 0. | | | 2.2600 | 0.9400 | 1.4000E 01D | 1.6966E-01 | 7.9743E-01 | 3.0453E 01 |
| | | | | | | | | 5.0000E 00Y | 1.5416E-01 | 7.2453E-01 | 2.7669E 01 |

WEIGHT OF CHAIN NO. 1 = 1.00 GRAMS

| POWER TOTALS | TIMES | W/GM | W/CC | W/KILOCURIE | CURIES |
|---|---|---|---|---|---|
| | 1.4000E 01D | 2.0674E-01 | 9.7168E-01 | 6.6100E 00 | 3.1277E 01 |
| | 5.0000E 00Y | 1.8695E-01 | 8.7865E-01 | 6.7582E 00 | 2.7662E 01 |

*Fig. I-8 Program ISOPOWER printout for $^{90}Sr$ isotopic power source.*

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Section

II

Isotopes and Radiation Technology

# Isotope Technology Development

## Sixth International Radiocarbon and Tritium Dating Conference*

By Edwin A. Olson† and Roy M. Chatters‡

*A summary of papers presented at the Sixth International Radiocarbon and Tritium Dating Conference in Pullman, Wash., June 7–11, 1965, is presented. Seventy-four papers were given at the conference on the following subjects: techniques for processing and counting of radiocarbon and tritium, archeologic applications in both Europe and the New World, geologic applications in both dating and hydrology, and general topics such as the correlation of radiocarbon dates to tree-ring chronology.*

Since 1954, workers with radiocarbon dating have met six times for international gatherings devoted solely to considering the assumptions, techniques, and applications of the age-determination method pioneered by Willard Libby. The sixth meeting was in Pullman, Wash., from June 7 to 11, 1965, and was attended by 145 conferees. Washington State University, whose Roy M. Chatters was general chairman, was host, and Edwin A. Olson of Whitworth College in Spokane served as program chairman. Through the financial support of the U. S. Atomic Energy Commission, the National Science Foundation, and the Wenner-Gren Foundation, the conference enjoyed a truly international flavor: 48 scientists representing 23 other countries presented slightly more than half the papers. W. F. Libby served as honorary chairman and keynoted the meeting with a luncheon address entitled Radiocarbon and Tritium in Retrospect and Prospect. For the first time papers on environmental tritium measurement were included because of a certain overlap with radiocarbon dating in both technique and application.

In addition to papers presented in technical sessions, special addresses were given by Thomas A. Rafter of New Zealand's Institute of Nuclear Sciences and Frederick Johnson of the R. F. Peabody Foundation for Archeology in Massachusetts. Rafter, one of the first workers to make natural radiocarbon measurements after Libby's initial publications, delighted his banquet audience with his talk, Problems in the Establishment of a Carbon-14 and Tritium Laboratory. After one of the conference luncheons, Johnson spoke to the group on The Impact of Radiocarbon Dating on Archeology. Few people appreciate this dramatic effect as well as Johnson, who was on the original committee of five called by Libby to obtain known-age samples for initially testing the $^{14}C$ dating technique.

In a specially called evening meeting, the conferees considered the question of the best half-life to use in reporting $^{14}C$ ages to the journal *Radiocarbon*. A similar discussion was held during the 1962 Cambridge Conference (5th on radiocarbon dating) as a result of three new measurements of half-life[1] that showed the accepted value of 5568 years to be 3% low. (The best available value for this half-life is now 5730 years.) Majority opinion[2,3] at both the

*The authors thank the following session chairmen for their written comments on papers presented in the sessions they moderated: Richard Daugherty (Washington State University), Arthur Fairhall (University of Washington), Gordon Fergusson (UCLA and Johnston Laboratories), Richard Foster Flint (Yale University), James Griffin (University of Michigan), Willard F. Libby (UCLA), Gote Ostlund (University of Miami, Florida), Meyer Rubin (U. S. Geological Survey), William Schell (Hazleton-Nuclear Science Corp.), Philip Smith (University of Toronto), and Eric Willis (Isotopes, Inc.).

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Cambridge and Pullman conferences was for retaining the old value for the sake of uniformity in publication but, at the same time, suggesting a correction factor of 1.03 to be applied for greater accuracy.

A highlight of the Pullman conference was an all-day field trip, which provided a change of pace in the middle of the conference week. The itinerary included the Palouse hills of Pleistocene loess deposits, the channeled scablands where Pleistocene flood waters have exposed and scoured Tertiary basalt flows of the Columbia Plateau, and the Marmes Rock Shelter at the confluence of the Palouse and Snake Rivers. The last has been excavated by Washington State University archeologists, who have exhumed several human skeletons antedating the Mazama ash fall of 6500 years ago. Guides for the field trip were Richard Daugherty (archeology), Roald Fryxell (geology), and James Crosby (geohydrology).

During the 4 days of technical sessions, 74 papers were presented. Highlights of the individual sessions are mentioned briefly here. For those who need more details, a limited number of copies of abstracts and proceedings are available from

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## Techniques

*RADIOCARBON PROCESSING AND COUNTING*

Since the previous radiocarbon conference, the closest approach to a breakthrough in $^{14}C$ techniques is the work of Tamers (Caracas) and of Noakes, Kim, and Stipp (Oak Ridge) in synthesizing benzene for liquid scintillation counting. They have simplified the chemistry and increased the yield to greater than 90% through two major steps: (1) using lithium to form the metallic carbide that yields acetylene on hydrolysis and (2) polymerizing the acetylene to benzene with a vanadium catalyst. Problems of isotopic fractionation, radon contamination, and reproducibility have apparently been solved. With the newer commercial liquid scintillation counters, background and count rates give a dating range comparable to the better gas-counting systems. In addition, automatic sample changing allows frequent background measurements.

Despite the promise of liquid scintillation counting, gas-counting techniques still dominate the field. Nydal (Trondheim) described a decade of experience using carbon dioxide; his techniques of gas purification, background reduction, and counter stabilization have continually improved through the years. Long (Smithsonian Institution) cited favorable experience with methane prepared by catalytic hydrogenation of carbon dioxide, and Fairhall (Washington) called attention to the difficulty of obtaining tritium-free hydrogen from commercial suppliers. Sharp (Sharp Laboratories) described his efforts to optimize counting characteristics in the methane counters produced by his company, placing emphasis on the solution of electronic problems such as noise and stability. Geyh (Hannover) uses ethane as the counting gas to introduce twice the amount of carbon into a liter-atmosphere of sample gas; by counting 24 liter-atm of ethane for 6 days, he can measure samples 60,000 years old without isotopic enrichment. Unfortunately a very small amount of sample contamination is very significant in this range and may make statistical calculations of range purely academic. A series of standard samples with ages extending from around 20,000 years, where $^{14}C$ ages should be reliable, back to 100,000 years or beyond is badly needed for evaluating contamination.

Finally, Oeschger (Bern) described a very small gas counter (40 $cm^3$ volume) designed to analyze carbon dioxide extracted from glacial ice. Even with so small a detector, almost 1 ton of ice must be melted to yield sufficient carbon dioxide for radiocarbon measurement.

*PREPARATION AND COUNTING OF TRITIUM*

In the measurement of natural tritium it is often necessary to enrich the $^{3}H$ prior to counting to attain adequate sensitivity. Customarily this has been done by water electrolysis; one such installation was described by Cameron and Payne (International Atomic Energy Agency, Vienna). Enrichment by thermal diffusion has lately been considered, and the conference heard reports on this technique from Sellschop (South Africa) and von Buttlar and Wiik (Darmstadt). Enrichment by a gas chromatographic column was described by Smith and Ahktar (Tennessee) but is not yet applicable to natural levels.

Isotopic enrichment is often unnecessary where tritium produced by a nuclear detonation

is sufficiently abundant, and proportional counting has been used without enrichment. Von Buttlar, Wohlfahrt, and Farzine (Darmstadt) generate hydrogen from natural waters and use it to hydrogenate inactive ethylene to ethane, which they count. Lal (Bombay) described a process to produce tritiated methane from water in one step. His reactor is loaded with sample water, zinc metal, and nonradioactive carbon dioxide gas; the net reaction is $CO_2 + 2H_2O + 4Zn \rightarrow 4ZnO + CH_4$. This same reaction can be used for $^{14}C$ measurements, in which case the carbon dioxide is the sample and the water is nonradioactive.

## Archeologic Applications

*RADIOCARBON DATING IN NEW-WORLD ARCHEOLOGY*

Since no group of scientists has used radiocarbon dating more than archeologists, three sessions were held in which the archeologic contributions, problems, and limitations of $^{14}C$ dating were outlined. Griffin (Michigan) focused his remarks on results in the eastern United States, where he noted the absence of satisfactory dating of many Archaic complexes and the conflict between archeological interpretations and radiocarbon dates for northern and southern Hopewellian cultures. He urged greater selectivity of samples for radiocarbon dating in the future. From Neuman (Smithsonian Institution) the conferees heard a report on archeology of the American Great Plains based on 214 dates from 121 sites, mostly representing hunting and gathering sites occupied from 9000 to 100 B.C. Other data show that the initial ceramic cultures date from around A.D. 50 to 700 and that agricultural settlements along the Missouri River and its tributaries fall between A.D. 700 and 1800.

In his report on the American Southwest, Jelinek (Michigan) described an attempt to correlate $^{14}C$ dating and palynology for the Cochise sequence in southeastern Arizona. He sounded an oft-repeated note concerning the lack of dates when he pointed up gaps in the dating record from 8900 to 7500 B.C. and from 4900 to 3300 B.C.; dates for ceramic cultures are also missing. Illustrating the dearth of $^{14}C$ ages is the fact that only 12 sites account for 60% of radiocarbon ages in the Southwest.

Haynes (Arizona) presented conclusions from a study of radiocarbon dates on sites of early man in the United States. Beginning about 11,500 years ago with Clovis points, he classified the available dates into 500-year intervals and traced the diversification of projectile-point types for 3500 years. Some points appear to be usable as index fossils. Such a use suggests that some $^{14}C$ dates from caves should be questioned. From his critical review he found that no reported $^{14}C$ dates older than 10,000 B.C. can be positively related to early man in the United States; he was impressed with the stratigraphic consistency of certain artifact types, fossils, and $^{14}C$ dates.

Three papers described archeologic work outside the United States. Borden (British Columbia) told of work he has done at three sites in the Lower Fraser Canyon near Yale, British Columbia, where a series of seven cultural phases dates from the eighth millenium B.C. almost to the present. He was able to relate human occupation to late Glacial geologic activity. Campbell (New Mexico) focused attention on the $^{14}C$ chronology of Arctic sites as related to chronologies based on other dating methods. In a paper on Peruvian archeology, Rowe (Berkeley, Calif.) described the problem of inconsistent results between $^{14}C$ dates made recently and those done in the mid-fifties. The later results move the time scale back about 600 years and suggest that the earlier values reflect difficulties with the obsolete black-carbon measurement technique.

*RADIOCARBON DATING IN OLD-WORLD ARCHEOLOGY*

In discussing dates from southwestern Europe and the Mediterranean Basin, Smith (Toronto) pointed out that the dating of the Mousterian sequence is not clear and that even in southwestern Europe the subdivisions of the Upper Paleolithic are not yet firmly dated, particularly in Spain and Italy. Judging from several $^{14}C$ dates from Libya, North Africa seems to have a surprisingly early form of Upper Paleolithic, and the lower Nile Valley was also less retarded than is usually thought.

Vértès (Budapest) discussed the evidence for the transition from Middle to Upper Paleolithic in eastern Europe, where available $^{14}C$ dates indicate a somewhat earlier transition than in western Europe; however, one cannot be sure it truly represents an independent development.

Vogel (Groningen) discussed the evidence that suggests some of the Mousterian and early

Upper Paleolithic dates from western Europe may be too young by several milleniums. He also introduced new evidence that the Paudorf oscillation extended from 30,000 to 26,000 B.C. and that the previously accepted Gottweig dates should be modified as well.

Bandi (Bern) discussed the Mesolithic age of western Europe, especially of Switzerland, and the evolution of the Epipaleolithic cultures in new environment. His results are based in part on his own work in aligning the cultural stages with late Glacial and early post-Glacial climatic history.

Clark (Cambridge) outlined the important contribution that radiocarbon dating has already made to an understanding of the diffusion of agriculture in the Old World, pointing out that the dates obtained have tended both to confirm the Middle East as the ecological focal area and to establish the priority of the Greek lands and the Middle Danube in the earliest spread into Europe. The dates have also expanded the time span of the whole European Neolithic period to the extent that important chronological subdivisions can now be established.

Waterbolk (Groningen), in describing the Copper Age of southeastern Europe, pointed out that in the light of recent radiocarbon dates we must revise our idea that copper working occurred earlier in the Iberian Peninsula than in southeastern Europe.

Moberg (Göteborg) emphasized problems of dating the many scattered unstratified sites of food-collecting peoples in Scandinavia during the last 5000 years. There is also great difficulty in correlating the chronology of northern Scandinavia with the better known southern Scandinavia sequence.

Agrawal (Bombay) presented a $^{14}C$ chronology for the Harappa culture, including its terminal phases, and for the first wave of Aryan-speaking people, represented by the Banas culture. The "Dark Ages" of India, between about 1500 and 500 B.C., are now being filled in, as Painted Gray-Ware sites are dated by radiocarbon.

*RADIOCARBON DATING: ARCHEOLOGY (GENERAL)*

A general session is admittedly one designed to catch all papers not fitting into established categories; yet in this session there was a common note of watchfulness in relating $^{14}C$ dates to archeologic problems. Stuckenrath (Pennsylvania) and Davis (Texas) sounded similar notes in their attempts to point up fundamental problems that may invalidate $^{14}C$ dates. They both emphasized the need for close cooperation between the collector and the laboratory personnel who make the radiometric analysis. In the connected fields of soil science and stratigraphic archeology, Dimbleby (London) described the potentialities and problems of radiocarbon dating. He suggested three specific applications: (1) turnover times in active soils, particularly in podzol B horizons; (2) time of change in soil character as a consequence of human or natural influences; and (3) the approximate age of buried paleosols.

Shutler (Carson City) mentioned problems unique to the tropics where heavy rainfall may cause contamination of samples. Where there are several internally consistent dates from a single site, there is reason for confidence. Shutler reported ages of earliest settlement in several Pacific islands, among which Sarawak showed the oldest age at 39,000 B.C. with the Philippines next at 20,000 B.C. From his series of ages, he inferred two possible migration routes outward from mainland Asia to the various Pacific islands.

Of particular interest was a paper by Oakley (British Museum) concerning $^{14}C$ dating of fossil hominids. He emphasized that there are really four distinct ways to determine the absolute age of fossil human remains: (1) from the material itself; (2) from associated materials; (3) from correlation with similar deposits dated absolutely; and (4) by inference on a largely theoretical basis. It was Oakley's contention that our knowledge of the age and relation of late Pleistocene hominids would be improved by a general recognition of these four methods of absolute dating.

## Geologic Applications

*TRITIUM AND RADIOCARBON IN THE ATMOSPHERE AND OCEAN*

For the most part this session concerned measurements of natural samples with heavy concentrations of $^{14}C$ and $^{3}H$ derived from the testing of nuclear devices. Although the dating aspect is not involved, much can be learned about mixing rates and turnover patterns in the atmosphere and ocean. This is so partly because both radiocarbon and tritium exist in the atmosphere as gases (carbon dioxide and water) in contrast to particulate fallout.

Fergusson (UCLA and Johnston Laboratories) presented recent stratospheric measurements for the Northern Hemisphere which showed maximum $^{14}C$ levels 10 times higher than the level at the earth's surface before nuclear devices had been exploded. A decrease in radiocarbon concentration occurs with decrease in latitude and altitude, although strong stratification is found in some sets of measurements. Additional stratospheric data for the Southern Hemisphere showed the familiar 18-month lag behind Northern Hemisphere values. For the troposphere, Lal (India) presented similar data measured in his country. Besides discussing the nature and time scale of north–south mixing in the troposphere, he included data on $^{90}Sr$ in rain to find out how fallout is washed from the atmosphere.

An original method of determining the source of water in the eye of a hurricane was described by Ostlund (Miami). Noting that rain accompanying 1964 hurricanes seemed depleted in tritium, he arranged for aircraft sampling of water droplets suspended at various points within hurricane Hilda. A more extensive sampling program is planned for the 1965 hurricane season, during which the extent of vortex interaction with the stratosphere will be investigated.

Recent ocean-surface measurements of both tritium and radiocarbon were presented by La Jolla scientists. Houtermans and Suess reported that 4 years of bimonthly sampling from 12 stations in the Pacific Ocean show that tritium concentrations are rising and that in the Northern Hemisphere values exceed those in the Southern Hemisphere. Radiocarbon measurements were given by Bien, Rakestraw, and Suess (La Jolla), who found evidence of a steady rise in concentration since 1954. The rate at which $^{14}C$ produced in nuclear explosions is taken up and mixed into the water masses of the deep ocean gives information on carbon dioxide exchange between the atmosphere and the ocean and, with tritium, provides evidence on rates at which surface water mixes downward through the thermocline.

Münnich (Heidelberg) closed the session with a comprehensive review of mixing processes in and between the atmosphere, ocean, and groundwater reservoir. He noted the inadequacy of simple box models to explain data that are becoming increasingly refined and then described convenient ways to handle more complex models. The illustrations he used concerned vertical mixing within the ocean and the atmosphere, where both $^{14}C$ and $^{3}H$ are useful, and soil water transport, where $^{3}H$ is more useful.

*CARBON-14 AND TRITIUM IN HYDROLOGY*

Growing interest in water-supply problems stimulated attention in this session in hydrological applications of $^{14}C$ and $^{3}H$. A dozen of the conferees gathered on two separate occasions to continue discussions begun in the session itself. Because of the difference in half-lives (5730 years for $^{14}C$ and 12.26 for $^{3}H$), both slow and fast water movements can be studied with these two radioisotopes.

Three well-known hydrological areas were topics for papers describing the use of radiocarbon in groundwater. Rubin, Hanshaw, and Back (U. S. Geological Survey) found increasing $^{14}C$ ages down the piezometric surface of the Ocala, Fla., limestone aquifer; their calculated rate of movement was 25 ft/year, in excellent agreement with estimates based on permeability methods. Similar agreement was found by Bennett (Arizona) for an artesian aquifer in Arizona, and studies are being extended to the Tucson Basin. Geyh and Wendt (Hannover) added stable-carbon-isotope measurements to radiocarbon data to study exchange processes between dissolved carbon dioxide and calcite within the aquifer.

Unlike the situation with radiocarbon, tritium produced in nuclear explosions is abundant enough today to dwarf natural concentrations of the isotope. Hence the use of tritium in hydrology requires many measurements today to establish the basis necessary for tomorrow's hydrological conclusions. The conference was told of the large body of excellent data now being accumulated routinely in a number of areas; for example, Thatcher and Payne (International Atomic Energy Agency, Vienna) and Stewart (U. S. Geological Survey) presented results gathered by their organizations. Both seasonal and year-by-year tritium variations were demonstrated.

Gat (Israel) described some of the problems and techniques in using tritium from nuclear detonations as a tracer for gaining insight into turnover times and mixing patterns of lakes and groundwater of semiarid Mediterranean regions. These include analysis, sampling, isotopic fractionation, and isotopic exchange.

*RADIOCARBON DATES IN GEOLOGICAL CHRONOLOGIES*

No single theme integrated the papers in this session aside from the application of radiocarbon dating to chronologic problems in geology. Dreimanis and Vogel (Western Ontario and Groningen) presented new dates and interpretations defining Wisconsin glacial fluctuations north of Lake Erie. Gonzales, van der Hammen, and Flint (Colombia, South America; Leiden; and Yale) described a $^{14}C$-calibrated pollen sequence found in lake sediments related to mountain glacier activity in the high Andes of Colombia. Their results support the view that major climatic events there were synchronous over the past 12,000 years with those in mid- and high-latitude North America and Europe. A similar time interval was the concern of Fryxell (Washington State), who integrated 50 $^{14}C$ dates into a post-Glacial chronology of the Columbia Plateau; two well-dated ash falls (Mt. Mazama at 6600 years B.P.* and Glacier Peak at 12,000 years B.P.) were especially useful stratigraphic markers.

The oölithic sands of the Bahama Banks were the topic of Martin and Ginsburg (Shell Development Co.). Chemical separation of these concentric spheroids into inner and outer fractions was followed by dating, results of which were consistent with radial growth. Since no apparent age older than 2700 years was found, a relatively rapid growth rate is indicated.

Berger, Ting, and Libby (UCLA) ended the session with a report on radiocarbon-dated materials—primarily coprolites—which are associated with pollen in caves of the southwestern United States.

## Other Topics

*TREE RINGS AND RADIOCARBON DATING*

Several times in conference discussions it was emphasized that laboratories do not measure ages; they measure sample activities. The connection between activity and age is made through a set of assumptions. However, in the session relating tree rings and $^{14}C$ dating, there was little need to emphasize this point because the session dwelt on one of the main assumptions of $^{14}C$ dating—that the atmospheric radiocarbon level has held steady over the age range to which the method applies. Tree rings have been selected as indicators of atmospheric $^{14}C$ levels in the past on the assumption that the carbon in a given ring reflects the atmospheric level when it was formed, provided a correction based on the $^{13}C/^{12}C$ ratio is made for isotopic fractionation.

Dendrochronologist Bannister (Arizona) opened the session by emphasizing the danger of universally assuming that a given tree ring can be dated merely by counting radially inward from the date of tree cutting. He outlined the effort needed to detect missing annual rings or multiple rings within a single year.

All other papers in the session reported on tree-ring analyses, many authors suggesting ways to explain deviations from the atmospheric uniformity assumed in $^{14}C$ dating. Schell, Fairhall, and Harp (Washington) analyzed the cellulose from *Sequoia gigantea* rings back to 700 B.C. Their results show a deviation that may be explained in one of two ways: either the half-life of radiocarbon is 5833 ± 127 years, slightly higher than the currently accepted value, or ocean temperature has linearly changed and hence gradually altered the ocean–atmosphere equilibrium relations and, in turn, the $^{14}C/^{12}C$ ratio in air.

Kigoshi (Tokyo) reported analysis of an 1800-year-old tree from Yaku Island in southern Japan. His results show a continuous decrease totaling about 2.5% during the past 1800 years. For 1500 years of this period, Kigoshi suggests correlation with measured variations in the earth's magnetic intensity as preserved in historically dated bricks. Such a magnetic variation would cause a variation in the cosmic-ray flux striking the earth's atmosphere and, in turn, would alter the rate of $^{14}C$ production.

Damon (Arizona) presented results on some of the oldest rings dated so far. He found a rise of 0.4% per 100 years in the atmospheric $^{14}C$ level from 500 B.C. to 2400 B.C., the latter date represented by rings from bristlecone pine. Egyptian archeological samples of the third millenium B.C. suggest the same high atmospheric $^{14}C$ level. In an effort to extend the sampling period still further, the University of Arizona Dendrochronological Laboratory has succeeded in correlating the outer rings of a long-dead bristlecone pine to the oldest part of the standard Southwest tree-ring chronology, thereby determining that an inner ring was some 5700 years old. Damon reported that the

*Before Present, defined as years before 1950 A.D.

atmospheric $^{14}C$ level indicated by this ring is 13% above that accepted as the standard used in age calculations and is equivalent to an age error of approximately a thousand years. A possible climatic element is Damon's suggested cause of atmospheric variation, but he presented no specific mechanisms.

Dyck (Geological Survey of Canada) presented results of dating Douglas fir rings spanning the past 1100 years. His results confirm previously published work. To explain the observed variations (which fall within a total range of 4.5%), he suggested that photosynthesis rates vary enough to account for short-term changes but that redistribution of carbon between the hydrosphere and the biosphere explains long-term changes. Both phenomenons presumably depend on climatic variations in temperature.

Stuiver (Yale) and Vogel (Groningen) both considered possible geographic variations of rings grown over the past several hundred years. For North America, Stuiver found uniformity regardless of latitude or elevation of the growth environment. Vogel's results for trees from Germany, Argentina, and the United States also agreed, provided corrections were made for isotopic fractionation. Both men measured with statistical errors of about 0.25%.

On the basis of 150 tree-ring measurements, Suess (La Jolla) suggested a correlation between the atmospheric radiocarbon level and sunspot frequency. He therefore postulated that atmospheric $^{14}C$ variations occurring with a period approximating 100 years are due to variations in the solar component of the cosmic-ray flux. When the time constant is of the order of 1000 years, the $^{14}C$ level of the entire cycle is affected; but so far there is no possible correlation of the existing tree-ring data with any relevant geophysical parameter.

*GENERAL SESSION ON RADIOCARBON DATING*

The first part of this session concentrated on problem materials submitted for age dating. Krueger (Geochron Laboratories) presented some encouraging results on the bone collagen remaining after other bone matter is dissolved in cold 0.5 *M* HCl. Although age diminishes the amount of collagen remaining in bone, some samples older than 40,000 years have yielded enough collagen for dating. Despite carbonate fractions that yield young ages owing to groundwater contamination, collagen fractions have consistently given what appear to be acceptable ages.

Mortar is another useful material, provided accurate dating is shown to be possible. If all the carbon in mortar is taken from the atmosphere during the hardening period, accurate dates should be obtainable. But the presence of $^{14}C$-free limestone in either the sand filler or the slaked lime will make measured ages too old. The standard empirical way to evaluate a given material is to date samples of known age. Such measurements carried out for mortars by two laboratories yielded opposite conclusions. Delibrias and Labeyrie (Gif-sur-Yvette, France) described their work and concluded that radiocarbon dating can be extended to mortars without any modifications. On the other hand, Stuiver and Smith (Yale) reported that all seven mortars they measured showed ages averaging 240 years greater than corresponding historical ages. Hence mortar must remain questionable material.

Tests of foraminifera derived from deep-sea cores were the subject of a paper by Olsson (Uppsala), who focused on potential contaminants. These she listed as (1) fine particulate limestone carbonate introduced through sediment reworking and (2) atmospheric carbon dioxide exchanged while a sample is in storage or being processed. Particle-size fractionation often isolates the first contaminant, and exchanged surficial carbon dioxide can be removed by acid leaching or thermal decomposition.

In a paper on groundwater $^{14}C$ ages based on dissolved carbonate analysis, Pearson (Texas) showed that satisfactory correction for the nonatmospheric limestone contribution was possible by use of a stable-carbon-isotope measurement. Without the correction, measured radiocarbon activities do not correspond to ages determined from hydrologic parameters; with the correction, agreement is good.

The latter part of the general session covered an assortment of topics. Libby (UCLA) pointed up the geophysical insights that might be possible from anomalies in radiocarbon dates—specifically, anomalies that appear to stem from variations in $^{14}C$ production rates by cosmic rays. He presented evidence that both solar and terrestrial magnetic moments have remained roughly constant over a time span measurable in tens of thousands of years. Olsson (Uppsala) presented a FORTRAN computer

program that her laboratory technicians use for $^{14}C$ age calculations during her absences from the laboratory. And Callow (National Physical Laboratory, England) made a plea for uniformity in the calculation and presentation of $^{14}C$ age results, especially with regard to errors quoted.

The final paper, by Thurber, Broecker, and Kaufman (Columbia), provided encouraging evidence that atmospheric uniformity over the past 25,000 years is approximately true despite the smaller scale variations brought out in the tree-ring session. They determined two distinct ages on a number of marine carbonates—one age based on radiocarbon and the second on the degree of inequilibrium between $^{230}Th$ (ionium) and its parent, $^{234}U$. There was agreement, within experimental error, over the range 3000 to 25,000 years. However, beyond 25,000 years, radiocarbon ages were much younger than corresponding ionium ages—a situation explained as resulting from $^{14}C$ contamination.

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Section

# III

Isotopes and Radiation Technology

# Food Irradiation

The food supply and the burgeoning population of the world are controversial, complex problems that are not easily solved. Some authorities believe that the distribution of tillable acreage and food supplies determines "have" and "have-not" regions—i.e., geography influences gastronomy.

The *supply* of food is essentially unaffected by preservation methods, but the *store* of food may be substantially increased by reducing

Table III-1 IRRADIATORS IN AEC FOOD PROGRAMS*

| Type | Purpose and status | $^{60}$Co gamma source | Capacity | Cost | Fabricator |
|---|---|---|---|---|---|
| Research irradiators, located at a number of educational institutions† | Immediate availability for research support; have been operating several years | 30,000 curies, double plaque, pool storage | 75 lb/hr at 1 Mrad | $35,000 plus source | BNL; Processing Equipment Corp., Lodi, N. J. |
| Mobile gamma irradiator, truck-mounted | Demonstration of fruit processing feasibility, economic determinations; was scheduled to begin operating in mid-1965 | 125,000 curies, four-pass quadrant irradiation | 1000 lb/hr at 200,000 rads | $225,000 plus source | Vitro Engineering Co, a division of Vitro Corp. of America, New York |
| Grain-products irradiator, USDA's Entomological Research Center, Savannah, Ga. | Bulk-grain or packaged-product disinfestation; was scheduled to begin operating in mid-1965 | 35,000 curies; stored in shipping cask, or extended to radiation positions for grain or packages | 5000 lb/hr bulk, or 2800 lb/hr packaged product, either at 25,000 rads | $200,000 plus source | Vitro Engineering Co, a division of Vitro Corp. of America, New York |
| Marine Products Development Irradiator, USDI's Technological Laboratories, Gloucester, Mass. | Semicommercial seafood irradiation; operating in 1965 | 250,000 curies, pool storage, four-pass quadrant irradiation | 1000 lb/hr at 500,000 rads | $500,000 plus source | Associated Nucleonics, Inc., Garden City, N. Y. |
| On-board ship irradiators, two transportable 18-ton self-contained units | Research support, operating in 1965 | 30,000 curies, continuous liquid irradiation | 150 lb/hr at 100,000 rads | $36,000 plus source | Nuclear Materials & Equipment Corp., Apollo, Pa. |
| Tropical fruit irradiator | Funds allotted in 1965; completion expected in spring 1967 | | | | |

*Adapted from *Atomics*, 18(3): 17 (1965).

†Universities of California (Davis), Florida, Hawaii, Michigan, North Carolina, North Dakota, and Washington and the Massachusetts Institute of Technology.

spoilage. In this way food irradiation can raise living standards and improve health. In any case political, economic, and social factors certainly need to be reckoned with in solving the food-supply problem.

Some of the benefits of food irradiation, such as savings resulting from reductions in both spoilage loss and the incidence of food-borne diseases, are also provided by other forms of food preservation (heat, drying, etc.). If food irradiation is to be adopted as a method of preservation, it should not only prove itself the equal of conventional preservation but should also demonstrate advantages over other methods. For example, is an irradiated product better than a dried one? Is it more feasible to install a food-irradiation plant in a developing country than to introduce conventional preservation methods or to influence the populace to change centuries-old habits? Is irradiation preferable to chemicals for inhibition of sprouting and for disinfestation of grains?

The AEC has sponsored a variety of irradiators (Table III-1). Of these, mobile and fruit irradiators appear to be particularly promising. A proposed and an actual installation are shown in Figs. III-1 and III-2. Both the mobile electron source, with its insulated-core-transformer (ICT) electron accelerator, and the fisheries-product irradiator illustrate irradiation at the site of harvest or catch—an important technique. Figure III-3 is a photograph of the Hawaiian Research Irradiator at the University of Hawaii, Honolulu, Oahu, which is to be used to evaluate the effect of irradiation of fruit in general and of tropical fruits in particular.

An important consideration in the development of methods is the possibility of combining conventional preservation methods with irradiation—perhaps with a synergistic effect. The two articles in this section include a discussion on the economic aspects of food irradiation and a review of the combination of food additives and irradiation for food preservation.

The economic aspects of food irradiation are significant factors in the acceptance of this method of food preservation. The U. S. Depart-

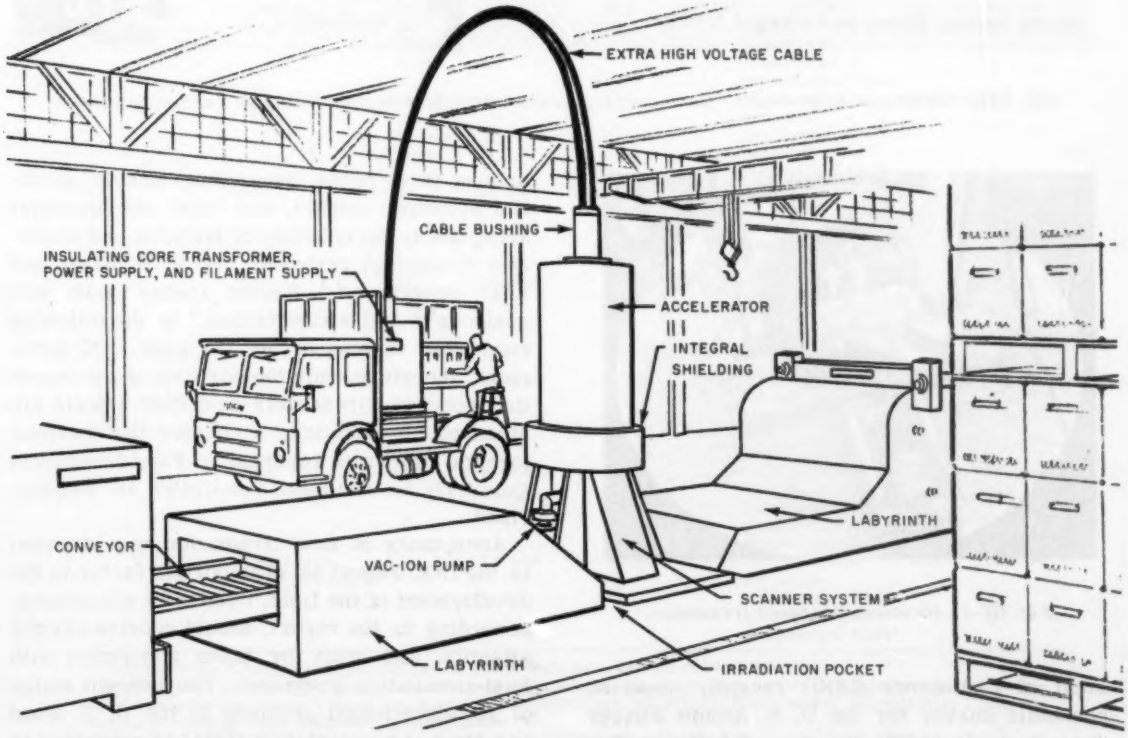


*Fig. III-1 Artists' conception of mobile electron source for food-processing system. Courtesy of High Voltage Engineering Corp., Burlington, Mass.*

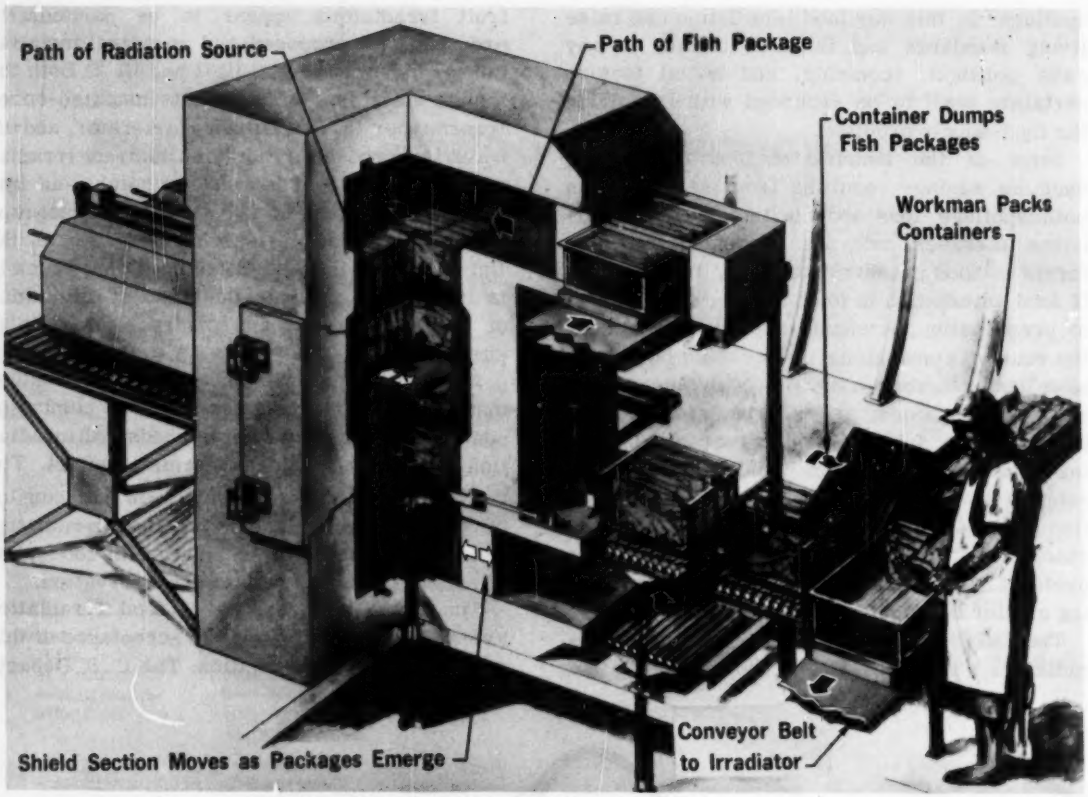
Path of Radiation Source
Path of Fish Package
Container Dumps Fish Packages
Workman Packs Containers
Shield Section Moves as Packages Emerge
Conveyor Belt to Irradiator

*Fig. III-2 Proposed large-scale $^{60}Co$ fishery-products irradiator installed on board a fishing vessel.*

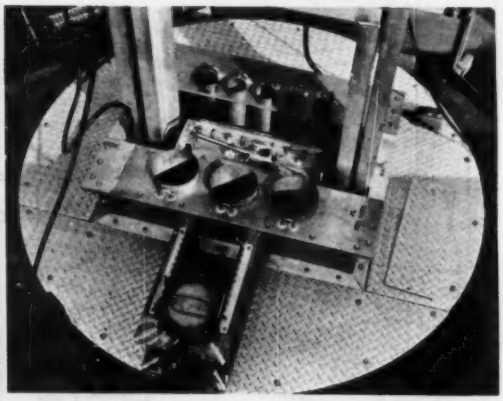

*Fig. III-3 Hawaiian Research Irradiator.*

ment of Commerce (DOC) recently made an economic survey for the U. S. Atomic Energy Commission's (AEC) Division of Isotopes Development (DID) and prepared a report[1] comprising three parts: general discussion, potential economic impact, and legal and consumer acceptance. Early issues of *Isotopes and Radiation Technology* reviewed the U. S. Army and AEC programs.[2,3] Recent issues dealt with seafoods[4,5] and strawberries.[6] In the following condensed version of the 174-page DOC document, therefore, only the portions of the report that concern foreseeable economic effects are stressed. In addition, a chapter that reviews foreign research reported in *Food Irradiation Quarterly International Newsletter* is summarized.

Acceptance of food irradiation was stressed in the DOC report as an important factor in the development of the field. Consumer acceptance, according to the report, should receive careful attention and study by those concerned with food-irradiation programs. The present status of irradiated-food petitions to the U. S. Food and Drug Administration (FDA) is presented in Table III-2.

Table III-2 CURRENT STATUS OF FOOD-IRRADIATION PETITIONS TO THE U. S. FOOD AND DRUG ADMINISTRATION*

| Product | Source | Energy, Mev | Permissible dose range, Mrads | Petitioner | Purpose | Date of filing (F) or approval (A) |
|---|---|---|---|---|---|---|
| Bacon | $^{60}Co$ | 1.17, 1.33 | 4.5 to 5.6 | Army | Sterilization | Feb. 8, 1963 A |
| | $^{137}Cs$ | 0.66 | 4.5 to 5.6 | AEC | Sterilization | Jan. 30, 1964 A |
| | Electron beam | 5 | 4.5 to 5.6 | GE | Sterilization | Aug. 23, 1963 A |
| | | 10 | 4.5 to 5.6 | Army | Sterilization | Apr. 15, 1965 A |
| | X rays from electron beam | 10 | 4.5 to 5.6 | Army | Pasteurization, sterilization | Dec. 15, 1964 A |
| Wheat and wheat products | $^{60}Co$ | 1.17, 1.33 | 0.02 to 0.05 | Brownell, Horne, and Kretlow | Insect disinfestation | Aug. 21, 1963 A |
| | $^{137}Cs$ | 0.66 | 0.02 to 0.05 | AEC | Insect disinfestation | Oct. 2, 1964 A |
| | Electron beam | 5 | 0.02 to 0.05 | High Voltage Engineering Corp. | Insect disinfestation | Dec. 12, 1963 F |
| White potatoes | $^{60}Co$ | 1.17, 1.33 | 0.005 to 0.010 | Army | Sprout inhibition | June 30, 1964 A |
| | | 1.17, 1.33 | 0.005 to 0.015 | Army | Sprout inhibition | Jan. 25, 1965 F |
| | $^{137}Cs$ | 0.66 | 0.005 to 0.010 | AEC | Sprout inhibition | Oct. 2, 1964 A |
| | | 0.66 | 0.005 to 0.015 | Army | Sprout inhibition | Jan. 25, 1965 F |
| Oranges | $^{60}Co$ | 1.17, 1.33 | 0.075 to 0.200 | Army, AEC | Inhibition of surface and subsurface microorganisms | Dec. 11, 1963 F |
| | $^{137}Cs$ | 0.66 | 0.075 to 0.200 | Army, AEC | Inhibition of surface and subsurface microorganisms | Dec. 11, 1963 F |
| Packaging materials | Gamma radiation | Depends on source, food | 1.0 max. | AEC | Food contactants for use in irradiation preservation of prepackaged foods | Aug. 10, 1964 A† |
| | Gamma radiation | Depends on source, food | 1.0 max. | AEC | Food contactants for use in irradiation preservation of prepackaged foods | Feb. 11, 1965 F‡ |
| | $^{60}Co$ | 1.17, 1.33 | 6.0 max. | Army | Food contactants for use in irradiation preservation of prepackaged foods | Dec. 8, 1964 F§ |
| | $^{137}Cs$ | 0.66 | 6.0 max. | Army | Food contactants for use in irradiation preservation of prepackaged foods | Dec. 8, 1964 F§ |
| Vegetable parchment paper | $^{60}Co$ | 1.17, 1.33 | 6.0 max. | Army | Food contactants for use in irradiation preservation of prepackaged foods | Mar. 8, 1965 A |
| | $^{137}Cs$ | 0.66 | 6.0 max. | Army | Food contactants for use in irradiation preservation of prepackaged foods | Mar. 8, 1965 A |
| | X rays from electron beam | 5 | 6.0 max. | Army | Food contactants for use in irradiation preservation of prepackaged foods | Mar. 8, 1965 A |

*Adapted from Ketchum, Osburn, and Deitch[1] and brought up to date as of May 1965.

†Nine classes of materials: nitrocellulose-coated cellophanes; glassine papers; wax-coated paperboards; polypropylene films; ethylene-alkene-1 copolymer films; polyethylene films; polystyrene films; rubber hydrochloride films; and vinylidene chloride–vinyl chloride copolymer films.

‡Polyethylene terephthal film, resins, and polymeric coatings for polyolefin film.

§Six classes of plastic materials: polyolefins, polyvinylidene chloride copolymers, polyvinyl chlorides, polystyrenes, polyesters, and polyamides.

# Potential Economic Impact of Commercial Radiation Preservation of Food*

*In this economic study 28 products were considered together with 6 radiation processes: sterilization, pasteurization, disinfestation, sprout inhibition, disinfection, and product improvement. Even though precise cost data are lacking, it appears that 17 of the treated products have "excellent" or "good" commercial prospects for domestic or international markets. Radiation holds the most promise at this time for pasteurized poultry, marine products, and strawberries; sterilized poultry and ham; improved dehydrated vegetables; and disinfected liquid, frozen, and dried eggs. Evaluations were based on such considerations as the state of technology, industry needs, estimated costs, competitive methods, and consumer habits and preferences. The impact of food irradiation on agriculture, fisheries, and food processing and related industries is assessed. The primary impact will be on individual segments of the market chain rather than on the food industry as a whole. Major benefits that may be expected to result from the commercialization of food irradiation include the following: (1) savings resulting from reductions in spoilage; (2) reductions in food-borne diseases and parasites; (3) market expansion and stabilization as a result of extensions in shelf life and shipping distances and of increased variety of products; and (4) expanded export potentials, especially in developing countries.*

During the past decade a great body of scientific knowledge has been developed about food and its reaction to ionizing energy. Many foods treated with ionizing radiation, either from isotopes or from electron machines, have proved wholesome, nutritious, and free of any measurable induced radioactivity. Consumers stand to benefit in a number of ways from the commercialization of food irradiation, which within the next decade should emerge as a major technique in food preservation. Commercial food irradiation also will affect economically those engaged in food production and distribution.

The major causes of food spoilage are changes brought about by the action of microorganisms and enzymes; chemical changes such as oxidation; physical changes such as dehydration; biological changes such as sprouting; and changes resulting from damage by insects, rodents, and other pests. As the food-distribution chain from producer to consumer becomes longer and more complex, food spoilage becomes a greater problem. At every step from the farmer to the retailer, the food industry wages war against food spoilage. The principal weapons in this war are canning, pasteurizing, cooling, freezing, dehydrating, pickling, fermenting, adding chemical preservatives, and carrying out pest-control programs. It is obvious that none of these principal weapons, or processes, preserves all foods and that no one food can be preserved acceptably by each of them independently. Some of the processes destroy certain vitamins and valuable nutrients, and others change the appearance, flavor, aroma, or texture of food. Considerable difference exists, for example, between a cucumber and a pickle, fresh and canned tomatoes, or grape juice and a grape wine.

*With the exception of thermal canning, the major food-preservative processes in commercial use today are merely sophisticated modifications of methods used by primitive peoples before the dawn of history.* [Editor's italics.]

It is not known exactly to what extent food irradiation may replace alternative methods of preservation. Food processors may adopt irradiation as a supplementary process, especially where its unique functions will fulfil needs not satisfied by other methods; on the other hand, a new and independent branch of the food industry may emerge composed of firms specializing in irradiation.

Food irradiation will affect agriculture, fisheries, food-processing and related industries, transportation, chemicals, marketing facilities, and world trade. Because of the relatively small number and volume of select food products that will be irradiated, however, compared to the tremendous size of the food and related industries, the primary impact will be on individual markets rather than on the food industry as a whole.

One of the most significant aspects of food irradiation lies in its potential for expanding international trade. Food irradiation can help to supplement the world's inadequate food supply, which must be greatly increased to feed a global population expected to double between the years 1960 and 2000. Concern about in-

*This article was condensed from a report by H. W. Ketchum, J. W. Osburn, Jr., and Jerome Deitch,[1] U. S. Department of Commerce. The work was done under a Division of Isotopes Development contract.

adequate food supplies has spurred food-irradiation research in many nations throughout the world. The recent union of the International Atomic Energy Agency (IAEA) and the United Nations Food and Agricultural Organization (FAO) in food irradiation activities underscores this concern.

## Measurement of Potential Economic Impact

Radiation processing of food, although the subject of a decade of research, experimentation, and development, has yet to be initiated commercially in the United States. In the absence of experience, the measurement of economic impact is subject to uncertainties and variables, such as:

1. Rate and success of technical research and development in radiation sources, facility design, and packaging
2. Development of acceptable cost data based on operations on a commercial or semicommercial basis
3. Determination of, on a commercial or semicommercial basis, benefits in spoilage reduction, extension of shelf life, improvements in quality, and market stabilization and expansion
4. Rate of issuance of regulations for radiation-processed foods by the FDA and, in some cases, the Meat Inspection Division and the Poultry Inspection Division, U. S. Department of Agriculture (USDA)
5. Extent of restrictive measures in federal, state, and local laws
6. Nature of consumer reaction and acceptance, and the success of consumer educational programs

## Food Production

The availability of food from farms and fisheries will undoubtedly be affected by the commercialization of radiation preservation of the specific products concerned. Both agriculture and the fisheries industry will have a large stake in the future developments of this new technology.

Table III-3 (compiled mainly from USDA sources) lists the 1963 production quantities and farm values of products most suitable for radiation processing. The fruit products listed accounted for over one-third of all fruit products in 1963, and their combined farm value was almost $600 million. Onions, potatoes, and tomatoes, which accounted for over 50% of all vegetables produced in 1963, had a farm value of approximately $675 million. Wheat production, which exceeded 68 billion pounds, had a farm value of $2.4 billion. Meat products that are likely candidates for radiation preservation (Table III-4) amounted to more than 37 billion pounds in 1963, with a retail value estimated in excess of $25 billion.

Table III-3 U. S. PRODUCTION OF SELECTED FRUIT, VEGETABLES, AND WHEAT IN 1963

| Food | Quantity, million pounds | Farm value, million dollars |
|---|---|---|
| Fruit | | |
| Apricots | 400 | 24.0 |
| Cherries (sweet and sour) | 304 | 40.9 |
| Figs (California) | 124 | 5.0* |
| Nectarines (California) | 114 | 5.3 |
| Oranges | 8,356 | 285.0 |
| Peaches | 3,536 | 137.0* |
| Strawberries | 510 | 94.9 |
| | 13,344 | 592.1 |
| Other fruit | 20,656 | |
| Total fruit | 34,000 | |
| Vegetables† | | |
| Onions | 2,522 | 85.3* |
| Potatoes | 27,600 | 441.6* |
| Tomatoes | 2,007 | 150.1 |
| Total | 32,129 | 677.0 |
| Other vegetables | 17,409 | |
| Total vegetables (including melons) | 49,538 | |
| Total fruits and vegetables | 83,538 | |
| Wheat | 68,280 | 2,390* |

*Estimated by Food Industries Division, DOC.
†Commercial production for fresh market.

It is generally accepted that radiation processing, to be most effective, should take place as soon after harvest or catch and as close to the point of origin of the product as possible. To the extent that field or mobile radiation may

Table III-4 U. S. PRODUCTION OF SELECTED MEAT AND POULTRY PRODUCTS IN 1963

| Product | Quantity, million pounds |
|---|---|
| Beef and veal (carcass weight) | 17,360 |
| Pork (carcass weight) | 12,360 |
| Bacon | 1,886* |
| Ham | 2,239* |
| Poultry | 7,410 |
| Chicken (ready-to-cook) | 6,070 |
| Turkey (ready-to-cook) | 1,340 |
| Total | 37,130 |

*Estimates by Food Industries Division, DOC.

prove feasible, this will affect the entire operation of handling and initial shipments of the crops involved. It is possible that mobile sources will be used in conjunction with permanent radiation processing sites provided with all necessary equipment except the radiation source. Such sites could be located in geographical areas of concentrated production for individual crops. A single mobile radiation source could serve a number of crops at different seasons or in different geographical areas. In some cases the permanent radiation sites might be owned and operated by individual large-farm owners, by farm cooperatives, or by other groups.

For many products, radiation processing probably will take place at central assembly points or at facilities operated by large food packers or processors. In these instances the producers will also benefit as a result of developments that increase the volume in marketings and receipts as a result of market expansion and seasonal stabilization. The elimination of all wastes due to spoilage and poor handling of fresh fruits and vegetables, for example, could reduce the acreage necessary for these products about 20%, and thus release nearly 1 million acres for other purposes.[7] Although this acreage is small in relation to the total of 300 million acres used in harvested croplands, individual farmers specializing in the production of fruits and vegetables would be significantly affected.

Table III-5 shows the volume and value of selected seafoods in relation to the total U. S. catch. The species selected are those for which favorable results have been achieved in radiation processing. The fishery products listed, data for which were compiled from Bureau of Commercial Fisheries sources, accounted for over 900 million pounds (17% by weight of the total 1962 catch) and had a value of more than $130 million (33% of the value of the total catch).

In 1960 the technical, economic, and practical aspects of radiation preservation of fish were evaluated by the Department of Food Technology, Massachusetts Institute of Technology,[8] and the marketing feasibility of radiation-processed fishery products was studied through a survey conducted by the Bureau of Commercial Fisheries, Department of the Interior.[9] These studies will be discussed later in this article.

Research on radiation pasteurization of fish has clearly demonstrated the feasibility of converting highly perishable fresh fish to a less perishable product by adding 20 to 30 days to the shelf life. This will make possible a leveling out of supply and demand in current seaboard markets and will permit a more orderly marketing process. It will also permit (1) the establishment of brand identity (because of market stabilization) as a means of increasing the volume of sale of quality products in major markets, (2) the creation of new markets for "fresh" seafoods in highly populated inland areas in which such products are not now available, and (3) more successful competition of U. S. seafood with imported seafood. As in the case of agricultural products, radiation preservation of marine products should take place as close to the point of catch as possible, preferably aboard ship.

It has been estimated that in the waters adjacent to this country about 7 billion pounds of fish annually remain unharvested; this represents a potential supply greater than the total present annual U. S. catch.[10] In addition, at least a billion pounds of the U. S. catch are now discarded at sea for lack of ready markets.

Table III-5 U. S. CATCH OF SELECTED SEAFOODS IN 1962

| Species | Quantity | | Value | |
|---|---|---|---|---|
| | Thousands of pounds | Percent of U. S. catch | Thousands of dollars | Percent of total |
| Flounder | 155,329 | 2.9 | 14,390 | 3.6 |
| Haddock | 134,250 | 2.5 | 10,913 | 2.8 |
| Ocean Perch | 141,310 | 2.6 | 6,046 | 1.5 |
| Pollock | 16,333 | 0.3 | 685 | 0.2 |
| Crabs | 243,340 | 4.5 | 18,708 | 4.7 |
| Shrimp | 191,106 | 3.7 | 73,236 | 18.4 |
| Clams (meat) | 54,169 | 1.0 | 11,762 | 3.0 |
| Total selected species | 935,837 | 17.5 | 135,740 | 34.2 |
| Balance of catch | 4,418,348 | 82.5 | 260,679 | 65.8 |
| Total U. S. catch | 5,354,185 | 100.0 | 396,419 | 100.0 |

The extent to which this potential for increasing the consumption of marine products may be realized through radiation preservation is contingent upon technical developments (which are highly advanced in the case of fishery products), the rate of legal clearances, and consumer acceptability. A study of consumer acceptability of irradiated marine products by the Bureau of Commercial Fisheries is currently in the planning stage and should help answer this question.

## Food Processing

The food-processing industry ranks first among all manufacturing industries in the United States in both value of shipments and total employment, and second in size of payroll. In 1962 the industry accounted for $67 billion, or about 7% of the value of shipments of all manufactured products, and employed 1.7 million workers, or 10% of all manufacturing employees, with a payroll of $8.6 billion. The types of food processors most likely to be affected by the development of radiation preservation include those currently involved in canning, freezing, freeze-drying, meat packing, fresh packing, and cold storage. Many firms in these fields are large and have adequate financial resources for launching commercial radiation operations.

For radiation pasteurization those firms now engaged in packing fish or fruits and vegetables might adopt the new technique for those products which are handled in large quantities. Firms dealing in freezing operations may expand into this field by using their knowledge of refrigeration techniques and their alliance with well-established refrigerated warehouses and distribution systems since, for maximum benefit, pasteurized products must continue to be refrigerated until ready for consumption.

Radiation sterilization, on the other hand, which requires no further refrigeration, may appeal to canners as a means of diversification since canners have the advantage of a long-time working relation with dry-grocery distributors through whom such products would logically be marketed. In addition, the present facilities of canners could be converted for irradiation by installation of an irradiation chamber at the point in the processing line where the product would otherwise have undergone thermal treatment.

Either the sterilization or the pasteurization of meat products could logically develop as part of the meat-packing operation.

Although the use of radiation to disinfest eggs and to tenderize dehydrated vegetables offers good commercial possibilities, petitions had not been filed with FDA for these items at the time of this writing.

## Packaging and Containers

Containers and packages are essential to the processing, distribution, and sale of all food products. The container and packaging industries meet the packaging needs of food processors and distributors, and they also represent a major consumer of many basic raw materials produced by other important industries.

The significance of packages and containers in the consumption of basic raw materials is illustrated by the fact that the packaging industry is the third largest customer of the steel industry, uses over 50% of the total production of paper and paperboard, and consumes approximately 90% of the aluminum-foil production. These figures relate to packages and containers for all types of products. Food is by far the most important of these products and accounts for nearly 60% of the value of shipments for the entire field; in 1962 the value of shipments of packages for food amounted to approximately $8 billion.

The impact of radiation preservation of food on those engaged in packaging will not be universal, nor will it be significant in total terms. Any changes will relate to particular types of containers and to the use of various types of packaging materials.

Studies have demonstrated that the packaging of foods to be treated with low doses of radiation (below 1 Mrad) does not involve serious problems.[11] Each product, however, has particular material requirements of tensile strength and moisture and vapor transmission.

By the Aug. 14, 1964, amendment to the food-additives regulations, FDA approved specified packaging materials for use in radiation preservation of prepackaged foods for doses not to exceed 1 Mrad.[12] The specified materials are nitrocellulose-coated cellophane, glassine paper, wax-coated paperboard, polypropylene film prepared from polypropylene basic polymer, ethylene-alkene-1-copolymer film, polyethylene film, polystyrene film prepared from styrene basic polymer, rubber hydrochloride film prepared from rubber hydrochloride basic polymer, and vinylidene chloride–vinyl chloride copolymer film. The basic research supporting the petition was conducted by the Hazleton Laboratories under a DID contract.

The U. S. Army Natick Laboratories have prepared an amendment to this FDA regulation for use in radiation preservation of prepared foods for six generic groups of packaging materials (polyolefins, polyvinylidene chloride, polyvinyl chloride, polystyrene, polyester, and

polyamide), which may be subjected to a sterilizing dose of irradiation not to exceed 6 Mrads for prepackaged foods.

Producers of packaging materials that are approved or currently under consideration in connection with radiation processing are well aware of developments in food irradiation and are interested in its possible effect. Many companies have either experimented in irradiating their own materials or have cooperated with other companies, the AEC, or associations to determine the effects of irradiation on the materials. Industrial representatives generally feel that any change in packaging requirements resulting from radiation processing can be met with little difficulty. The industries have large capacities, and, although they are operating considerably below capacity, they are growing at a substantial rate.

The effect of radiation preservation of food on the use of tin plate, aluminum plate, and metal foils will reflect primarily the success in the field of radiation sterilization. A major objective of the U. S. Army's Radiation Preservation of Foods Program is the development of flexible containers to replace metal cans for radiation-sterilized foods. Since radiation sterilization does not involve intense heat or pressure, flexible packaging can be used to reduce weight and shipping space. The need for effective and acceptable flexible packaging, including pouches, of plastic materials or laminated foils offers a major opportunity for producers of such materials.

## Radiation Sources and Facilities

During the past decade a radiation-processing industry has developed for the treatment of a number of important products in areas other than food irradiation. It is estimated[13] that the current annual sales of radiation-processed products exceed $70 million and are growing rapidly.

Sources of radiation for food preservation that presently appear to be most promising are gamma-emitting radioisotopes (primarily $^{60}Co$) and high-energy electron accelerators. Gamma rays from $^{137}Cs$ and X rays from electron-beam conversion are being considered.

Cobalt-60 food irradiators currently in use, under construction, or being designed for use with the AEC food-irradiation research program include research irradiators, transportable or mobile irradiators, central or in-plant irradiators, bulk-grain irradiators, and shipboard irradiators. Present costs for commercial irradiators are considered high in comparison with future costs since these initial investments reflect a higher proportion of design expenses.

A number of electron-accelerator facilities have been used in food irradiation, including a 24-Mev 18-kw linear accelerator now in operation at the U. S. Army Natick Laboratories. According to estimates by a manufacturer of radiation facilities, the 1964 sales of electron machines alone surpassed in kilowatt equivalent the sales of both electron machines and isotope sources combined for the previous 10 years.

The commercialization of radiation processing of foods will generate a demand for food-irradiation facilities. Mason and Taimuty[14] projected the potential for radiation-processing equipment for food preservation to the year 1980. According to this projection, in which it was estimated that 10% of the market for a number of foods would comprise foods processed by radiation, 20 billion pounds of radiation-processed food with a value of $4 billion would be consumed annually. More than 300 large accelerators would be needed for this volume, and the development would mean an annual market of about $15 million for the electronics industry.

As part of the demand for electronic supplies, control instruments and related components will be required. On the basis of present costs of suitable instruments for radiation facilities, total instrument costs for food-irradiation facilities of all types are expected to fall within a range of $15,000 to $35,000 each, depending on size and characteristics of the installation. Thus, should the demand for food-irradiation facilities reach a modest estimated level of 50 units per year, the annual market for control instruments would be approximately $1 million to $1.5 million. This represents less than 1% of the current total annual factory shipments for such instruments.

The evaluation of such projections necessitates careful analysis of the percentage of each product category which might ultimately be treated by this process, the type of treatment, the size and the source levels of facilities to be employed, the number of days and shifts per day of operation, the efficiency of source use, maintenance, and other variables.

## Refrigeration and Storage Facilities

The need for food refrigeration extends from the farm to the kitchen, and involves process equipment, transportation cooling devices, refrigerated storage facilities, refrigerated sales equipment, and finally, home refrigerators and freezers and refrigeration installations for restaurants and institutions.

The fraction of the retail sales dollar, over one-half, spent in food stores for products requiring refrigeration emphasizes the importance of commercial refrigeration. Approximately $250 million is invested each year in refrigerated equipment in retail food establishments.

Most fresh and perishable foods are refrigerated. Radiation-pasteurized foods will still require refrigeration. Hence any displacement of fresh foods by radiation-pasteurized foods will not reduce, but will more than likely increase, refrigerated space. This increase will result from longer holding periods, greater length of hauling distances, expanded geographical markets, and increased varieties of perishable products (e.g., tropical fruit) shipped into new markets.

The impact of commercialization of irradiated foods on refrigeration at the retail level will be diverse but will present no problems. The need for refrigerated display cabinets should increase with an increased volume of radiation-pasteurized products requiring refrigeration. On the other hand, there will be some reductions in refrigeration requirements for meat and produce with the shift of packaging from the retail outlet to the process station.

Commercial availability of radiation-sterilized foods may cause the rate of growth of refrigeration requirements to be slightly curtailed since such foods will require no refrigeration subsequent to treatment. However, no early shifts of this type are anticipated in connection with the sale of radiation-sterilized products in domestic markets since refrigeration facilities for both shipment and distribution are adequate and since the extension of shelf life for such products is less significant to consumers than it is for products likely to be pasteurized. A notable exception to this conclusion is the domestic sale of products such as sterilized canned hams, brown-and-serve turkeys, and roasts. In addition, large-scale food distributors have indicated an interest in the availability of radiation-sterilized meats, luncheon meats, bologna, and frankfurters to reduce their requirements for refrigerated food-display cases.

In foreign markets, especially in the less developed nations where refrigeration is inadequate, radiation sterilization may contribute to meeting consumer needs without creating a sudden demand for refrigeration facilities (which are inadequate or not presently available). Thus the use of sterilized foods may even retard future development of refrigeration facilities in these countries.

## Transportation

Food is undoubtedly the most important single commodity contributing to the transportation industry and its growth. Nevertheless, commercialization of radiation preservation of food will have only a relatively small and gradual effect in this industry within the foreseeable future. The effect on transportation facilities and requirements will be reflected in (1) refrigeration, (2) length of haul resulting from extension in shelf life, market expansions, and increase in the value of shipments, (3) geographical shifts in movements resulting from modifications in the location of assembly, processing, or storage points, and (4) competitive shifts in the proportion of various products handled by different types of carriers. Irradiated foods can be shipped farther by slower forms of transportation than highly perishable unirradiated products. This factor appears to favor increased volume by rail and trucklines over airlines. All types of carriers should benefit from a more orderly scheduling of shipments with relatively less equipment in relation to the total volume of shipments. Increased volume should result from shipments into markets not previously served; for example, large shipments of fresh ocean fish.

Shippers engaged in foreign trade should benefit significantly from increased exports of the longer shelf-life pasteurized perishable products and market expansion for sterilized products in areas where refrigeration facilities are inadequate. Shifts in competitive advantages and rates of growth may occur as a result of a change in the tonnage of specific products handled.

## Chemicals

The magnitude of the effect of radiation processing on chemicals cannot be determined now. Any significance that the commercialization of radiation processing of food may have on the chemical industry will relate primarily to the use of plastic materials in food packaging and the possibility of declines in the demand for pesticides, sanitizers, sterilizers, and sprout inhibitors. In addition, the use of chemicals as food additives and as refrigerants also may be affected.

## Marketing and Distribution Facilities and Operations

Food marketing and distribution is the largest business in the world. In 1963 American consumers spent approximately $76 billion for food (excluding alcoholic beverages), an amount that represented approximately 20% of all personal outlays.

There has been no comprehensive research concerning the probable impact of radiation processing of food products on marketing operations and practices at the wholesale and retail levels. However, in a survey[9] on the marketing feasibility of radiation-processed fishery products, the producers, wholesalers, and retailers indicated that the process would completely revolutionize the fresh-fish industry. In another study[15] on fresh fruits, the majority of respondents (two-thirds in the wholesale and retail segments) thought that the irradiation process would increase the production and market volume of the products covered. They did not foresee, however, a significant change in output and sales volume of canned, frozen, or other processed foods.

Changes in marketing activities will be less pronounced for radiation-pasteurized products than for sterilized products. It is possible that pasteurized perishable products will channel through fewer and larger shippers as a result of the volume requirements essential to economical processing costs. Since most irradiated products are prepackaged, there could be a significant shift in the packaging function from the retailer to the point of radiation processing. This shift could reduce both equipment and labor requirements at the retail level, providing savings that should more than offset the increased costs at the processing level.

At the retail level extended shelf life in some instances may reduce or help alleviate procurement problems, improve inventory control for the weekly sale cycle and unusual weekend carry-over, and reduce the need for distress sales. Shippers, at the same time, can anticipate increased flexibility in the selection of markets.

Little or no impact is foreseen in the distribution of disinfested flour and bakery products. Some changes may take place in the location of assembly, storage, and shipping facilities for grains. Similarly, for radiation processing of potatoes and onions for sprout inhibition, little marketing effect is seen beyond the point of application except for the effect of market stabilization by increased holding periods.

Radiation preservation of foods will not increase food-distribution and -production costs or processing costs except those associated with the radiation processing itself. To the contrary, substantial savings will accrue during the distribution of irradiated foods because of a reduction in refrigeration requirements for sterilized products and in spoilage losses for both sterilized and pasteurized products and because of market stabilization and expansion.

## Significance of Irradiated Foods in World Trade

The latest World Food Budget,[16] with projections through 1970, prepared by the Foreign Regional Analysis Division, USDA, pointed out that two-thirds of the world's people live in countries with nutritionally inadequate diets.

Radiation preservation of food offers great promise of contributing to the solution of current and emerging world food problems by adding to the total effective food supply through the elimination of losses due to spoilage, infestation, and sprouting. In addition, it will make possible the movement of foods whose present keeping qualities are inadequate for distant shipping. The importance of food as a factor in world trade is indicated by the amount of food and feed products exported in 68 major countries in 1961: $18 billion out of $105 billion for all products exported. It is anticipated that the effect of food irradiation on foreign trade will be most significant for grain, meats, fruits, poultry, and fish. In addition, significant export markets may also develop for radiation-processing facilities and equipment.

The losses in tonnage and dollar value sustained because of spoilage and insect infestation of grain are large. The consequences of these losses are reflected not only in reduced profits but also in a poorer diet for the peoples of the grain-deficient countries of the world—South America, Europe, Asia, and Africa—to which the bulk of such exports is directed.

Exports are an extremely important part of the billion-dollar poultry industry in the United States, which commands approximately one-half of the world's trade in poultry products. American poultry producers have a competitive advantage in costs and quality compared with other countries from both advanced technology and the economies of large-scale mass production. Frozen chickens, broilers, and turkeys, exported from the United States, because of their high quality, compare favorably in foreign markets with fresh poultry produced domestically. If the American preference of eight to one for fresh chicken in comparison with frozen chicken is considered, the foreign-market potential for fresh irradiated U. S. poultry could be substantially greater than the current export volume of frozen poultry.

In countries with inadequate refrigeration, only dried, smoked, or canned seafoods are available today. The possibility of providing these areas with sterilized fresh marine products that do not need refrigeration is highly significant.

In the United States, fish, unlike poultry, is imported extensively ($475 million in 1963) and accounts for approximately one-third of world fishery imports. A number of countries, notably Canada, Germany, the Soviet Union, and Japan, are at present successfully operating factory ships on which fish are caught and processed. The addition of on-board ship irradiators would result in a rapid and economical means of providing pasteurized packaged fresh fish for direct shipment to importing countries.

Although importers of grain and grain products, the countries of South America, Asia, and Africa are exporters of fruit. The development of an internal radiation capability would increase their exports of fruits and other products, which would, in turn, increase their ability to purchase grains and equipment. For example, food exports such as coffee, cocoa, and bananas are the basis of much of the Latin American economy. These crops all suffer losses through spoilage. The incidence of mold because of the high moisture content of coffee might be decreased by irradiation. In addition, many fruits cannot be shipped in international trade because of perishability or quarantine barriers. These fruits could become likely prospects for international trade if successfully irradiated. Reduction of spoilage and extension of shelf life for tropical fruits would also help correct the diets of the people of these countries.

A number of developing nations have already indicated an interest in facilities for food irradiation. In these countries food preservation is a much more important consideration than it is in more-developed nations. In the food-deficient areas of the world, the problem is not alone one of the unproductivity of the land or oceans. It is more apt to be the high perishability of domestic protein and vitamin sources and the tremendous wastes involved owing to lack of adequate processing, storage, transportation, refrigeration, and marketing facilities.

Both the import of irradiated foods and the irradiation of domestically produced foods will be a definite step toward solving the food-supply problem in many areas of the world and will contribute to an increase in living standards.

## Summary of Major Foreign Research

In the 18 countries belonging to the Organization for Economic Cooperation and Development (OECD), 12 European laboratories are working on irradiation of meat, 9 on the effects of radiation on seafoods, and 20 directly or indirectly with fruits and vegetables. A number of other countries are studying the effects of radiation on basic food elements. The total effort is modest, and not many more than 100 workers were engaged in research in 1962. [In 1960 the European Information Center for Food Irradiation (EICFI), located at the National Institute of Nuclear Science and Technology, Saclay, France, was established to keep researchers abreast of work in the field of food irradiation.[17]]

In general, experiments on varieties of fruits and vegetables indigenous to each country have been rewarding. In Canada potatoes were successfully irradiated and marketed.[17a] On the other hand, Russia, the first country in the world to issue a public clearance for the use

of ionizing radiation on food (1959), has been working on potato sprouting since 1954 but has made no commercial application.[18] Other work with potatoes is being done in the Republic of South Africa, Denmark, Australia, India, and Poland. Table III-6 gives a sampling of recent work done by OECD countries.

Recently IAEA and FAO united their activities in the application of isotopes and radiation to the food and agricultural sciences. Their program includes placing irradiated food in international commerce, disinfestation, control of harmful organisms transmitted by products moving in international trade, quarantine control by irradiation, sensitizing microorganisms to radiation, and conducting an international fruit-juice program. Research also will continue on marine, meat, and fruit products and their improvements in marketability.

Table III-6 HIGHLIGHTS OF MAJOR FOREIGN RESEARCH IN FOOD IRRADIATION[1,17b]

| Country | Foods and areas of study |
|---|---|
| Austria | Fruit juices, wines, beer |
| Australia | Control of Queensland fruit fly[17c] |
| Belgium | Strawberries[17d] and vegetables, grain, flour, powdered eggs, gluten; disinfestation of grain and grist |
| Denmark | Consumer tests planned on meats; potato sprouting; strawberries[17d] and other fruits and vegetables |
| France | Tomatoes, chicken, fermented milk, grain,[17e] economic study on irradiation of African produce (disinfestation of fish, sterilization of meat) |
| Germany | Basic studies of dosimetry in foodstuffs and effects of radiation on microbes; packaging; potato sprouting; fruits and fish[17f] |
| Greece | Radioresistance of yeasts and molds in fruit; disinfestation of figs; extension of shelf life of Valencia oranges[17g,h] |
| Holland | Pilot plant for strawberries planned[17i] |
| India | Mangos, sapotas, guavas, green tomatoes, limes, ginger, garlic, and beans[17j] |
| Israel | Irradiation of plastic-coated oranges[17k] |
| Italy | Inactivation of foot-and-mouth virus in imported beef |
| Norway | Marine products, beef, potatoes, fruit, and vegetables |
| Pakistan | Disinfestation of leguminous pulses[17l] |
| Poland | Potato sprouting; fruit, barley, and malt; mushrooms, cellulose, milk, meat; bilberries in polyethylene; development of potato decoctions for forage[17m] |
| Republic of South Africa | Apples, grapes, peaches, juices[17n] |
| Sweden | Wholesomeness of potatoes; fresh and smoked fish |
| United Kingdom | Wholesomeness of wheat, eggs; studies of Salmonella in horse meat for pet food; fish; strawberries[17o,p] |
| USSR | Potato sprouting; irradiation of hen eggs in hatchability and egg-production studies; extension of shelf life of several foods; disinfestation |

In furthering this program the Joint IAEA–FAO Division of Atomic Energy in Agriculture will support fellowships, training courses, exchange professors, research contracts, consultants, symposia and conferences, and technical assistance. They will also act as executing agent in programs funded by other organizations such as the United Nations or World Bank.[19]

(*Editor's Note: Since the appearance of the DOC report, two recent issues of* Food Irradiation Quarterly International Newsletter *have reported on further progress in OECD food-irradiation programs,[20] preservation of Venezuelan vegetables,[21] and studies in Japan.[22] In Japan several research institutes and universities, in addition to studying theoretical aspects of food irradiation, are sterilizing shell fish, paste products, and brewed foods; pasteurizing haddock; and inhibiting sprouting of potatoes and chestnuts.*)

# Combination Processes in Food Irradiation: Use of Food Additives

By J. S. Lee, D. A. Corlett, Jr., M. A. Shiflett, and R. O. Sinnhuber*

*Chemical agents, which may be classed as antibiotics, radiation sensitizers, and chemical preservatives, were investigated for possible use with food irradiation. The net result of such combinations is more than the individual additive effect, and often a distinct bacteriostatic or bactericidal action results. The current hypothesis for the mechanism, as well as the feasibility of combining irradiation with addition of chemicals for preserving foods, is discussed.*

The radiation dose that can be applied to food is often restricted by the flavor threshold, and both physical and chemical means of augmenting the radiation effect have been sought. Increased effectiveness is desirable both with low-dose irradiation, often called radiation "pasteurization," which must be combined with low-temperature storage of the product, and with high-dose irradiation, or sterilization. The net result of a combined process, whether physical or chemical, is to obtain an effect equivalent to that of increasing the radiation

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dose. Such processes, therefore, would permit lower radiation doses for some radiation-sensitive foods, would add to the already growing list ot foods that may be preserved by radiation, and would increase the margin of safety for irradiated foods.

Among the physical treatments that show promise are mild heating prior to or during radiation exposure, exclusion of air by vacuum packaging, and irradiation of food in a frozen state. Discussion of physical modifications, however, is beyond the scope of this paper, and the interested reader is referred to an excellent monograph by Goldblith.[23]

Combination processes using chemical agents can be divided into two classes. In processes of the first class, the agent is intended to increase the efficiency of the radiation against all microorganisms, thereby giving an effect similar to that of a higher radiation dose. In the second class, certain food additives are sought to eliminate a specific group of microorganisms that may not be inactivated by low-dose irradiation (e.g., clostridia spores). Most information available is for agents of the first category, about whose selectivity little is known.

The chemical agents discussed in this paper are classified as antibiotics, radiation sensitizers, and chemical preservatives.

## Chemical Agents

*ANTIBIOTICS*

The tetracyclines are the antibiotics most extensively studied for increasing radiation effectiveness. Most antibiotics are particularly potent against Gram-positive bacteria. On the other hand, low-dose irradiation alone eliminates most Gram-negative pseudomonads in foods. Therefore the process combining antibiotics and radiation is intended to cope with the Gram-positive bacteria that might survive irradiation.

Niven and Chesbro[24] first reported that spoilage of red meat could be delayed by 10 ppm of chlorotetracycline (CTC) and 500,000 reps of gamma radiation. The eventual spoilage of their samples was caused by yeasts, and the addition of 1000 ppm of potassium sorbate was advocated to prevent yeast growth. Phillips et al.[25] observed that 7 ppm of CTC and 200,000 rads of gamma radiation were necessary to obtain a significant storage-life extension of beef. Cain, Anderson, and Malaspina[26] similarly observed that CTC could enhance the effectiveness of radiation but also noted a rapid destruction of CTC and oxytetracycline (OTC) as a result of radiation exposure.

Ingram and Thornley[27] used CTC to increase the storage life of irradiated chicken. In chicken, however, the predominating flora changed from pseudomonads to achromobacteria with irradiation doses between 125,000 and 250,000 rads. Apparently CTC was as effective against achromobacteria as it was against Gram-positive bacteria.

Lerke, Farber, and Huber[28] studied OTC for extending the storage life of irradiated fish. They found that a combination of 5 ppm of OTC plus 11,800 rads of beta radiation produced a result comparable to that of irradiation with 280,000 rads for low-fat fish. The medium- and high-fat fish did not respond as well to this treatment. Increasing the OTC concentration to 20 ppm, however, gave comparable results with high-fat fish. The complementary effect of OTC and radiation on fresh shrimp was reported by Awad, Sinnhuber, and Anderson.[29] Chemical and flavor evaluations indicated a three- to fivefold extension of the storage life at 38°F. The use of OTC and CTC at a level of 5 ppm is permitted[30] in the United States in fresh (uncooked and unfrozen) fish, scallops, and shrimp.

Recently a new antibiotic, tylosin, has attracted attention. Greenberg and Silliker[31] have shown that it is active against the freshly germinated cells of spore-forming bacteria. This property is very attractive for food use since, if a contaminated food is to become toxic, the spore must be able to germinate and grow in food. Since tylosin attacks the early vegetative cells, the potential toxin producer may be inactivated before any toxin can be produced. Malin[32] was able to prevent the outgrowth of 50 strains of *Clostridium botulinum* types A and B and 10 strains of *Bacillus* species with tylosin concentrations ranging from 0.1 to 1.0 ppm in the medium. *Clostridium botulinum* type E also appears to be sensitive to tylosin. Sheneman[33] incorporated 100 ppm of tylosin lactate in brine and treated smoked whitefish chubs with this solution and heat. The sample did not produce toxin lethal to mice, but the controls did. Malin, Dobbs, and Martin[34] and Abrahamsson[35] further demonstrated the potential application of tylosin to decrease the hazard of *C. botulinum* type E in processed fish.

There is little information on the possibility of using tylosin in combination with radiation, but Poole and Malin[36] showed that tylosin activity diminished rapidly upon standing. If tylosin is stable under irradiation, it can probably be used to inhibit spore formers in irradiated foods.

*RADIATION SENSITIZERS*

Bridges and Horne[37] defined the radiation sensitizer as a chemical agent in whose presence the microbicidal effect of irradiation is greater than the sum of the effects of irradiation and of the agent separately. The sensitizer often has no activity by itself, and it must be present during the radiation exposure. In most cases sensitization has been observed only under anoxic conditions. These properties of sensitizers are valuable for radiation food sterilization where complete inactivation of all microorganisms is demanded. Proper use of a sensitizer allows the sterilization dose of radiation to be decreased, and vacuum-packaged foods can be irradiated with added confidence because of the anoxic nature of sensitization.

There are many proved radiation sensitizers. At present, however, only vitamin $K_5$ appears to have any possibility for food use. El-Tabey Shehata[38] and Silverman, El-Tabey Shehata, and Goldblith[39] reported that up to 40 ppm of vitamin $K_5$ and its analogs 4-amino-1-naphthol and 2-amino-1-naphthol decreased the survivors of strains of *Bacillus subtilis* and of *B. stearothermophilus* irradiated in a broth under a nitrogen atmosphere. The radiation-sensitizing effect of vitamin $K_5$ is not limited to these spore formers but has been seen in *Micrococcus radiodurans*, a highly radiation-resistant bacterium isolated from irradiated meat by Anderson et al.,[40] and in *Streptococcus faecalis* and *Escherichia coli*. The potential for the food application of vitamin $K_5$, however, appears to be less promising than the above-observed effects in buffer. When microorganisms were irradiated in orange juice or in milk, no sensitization with vitamin $K_5$ could be demonstrated. Even in buffer, 28 ppm of cysteine antagonized[39] the sensitizing effect of vitamin $K_5$.

*CHEMICAL PRESERVATIVES*

The simple preservatives sodium chloride, sodium nitrite, potassium nitrite, sodium nitrate, and potassium nitrate have been used in pickled meats, hams, and comminuted meats. Yesair and Cameron[41] reported a 70% decrease in the spore count of *C. botulinum* by 0.1% sodium nitrate, 0.005% sodium nitrite, or 2% sodium chloride. Bulman and Ayres[42] also found that nitrites were effective against the putrefactive anaerobe 3679. In studies dealing with these additives and radiations, Krabbenhoft et al.[43] reported that *C. botulinum* type A spores could be inactivated in inoculated meat by a radiation dose far below that required to obtain complete sterilization. *Clostridium botulinum* type A spores, inoculated in ground beef to give $10^6$ viable spores per gram, were completely inactivated by 2.0, 2.5, or 3.0 Mrads when the meat was treated with 2.5% sodium chloride in combination with either 1000 ppm of sodium nitrate or 200 ppm of sodium nitrite. The spores were not inactivated by radiation alone or by the additives alone, and, when not combined, the additives did not inactivate the spores after radiation exposure. Kempe and Graikoski[44] noted that curing ingredients and low-dose irradiation inactivated *C. botulinum* types A and B in pork luncheon meat. Segner, Schmidt, and Boltz[45] showed that *C. botulinum* type E in fish substrate was inhibited by sodium nitrite. They also reported that the outgrowth time of *C. botulinum* type E in fish homogenate was delayed when 0.1% sodium benzoate was added. Lee, Shiflett, and Sinnhuber[46] examined the combination of low-dose irradiation and sodium benzoate to increase the storage life of fish fillets. Sodium benzoate was not expected to have any antimicrobial effect at or near neutral *p*H, and Tarr[47] showed that benzoic acid and its salts were not effective preservatives for fresh fish due to their neutral *p*H. Bosund[48] attributed this effect to benzoate not being able to penetrate into microorganisms at the neutral *p*H.

Our experimental findings[49] showed that dover sole fillets remained at *p*H 6.8 after irradiation. Despite this, when either the fillets were irradiated in the presence of 0.1% sodium benzoate or the benzoate was added after radiation exposure, the cold-storage life of the fillets was approximately doubled at radiation levels between 0.1 and 0.5 Mrad. Furthermore, equal concentrations of potassium sorbate and sodium salts of esters of either methyl- or propyl-*p*-hydroxybenzoic acid all showed similar effects in retarding microbial growth in irradiated fillets.

## Mode of Action

There are several interesting similarities, as well as differences, among the agents discussed above. Some considerations for the mechanisms involved in these actions are of theoretical as well as practical importance. Radiation sensitization was recently discussed by Eldjarn and Pihl,[50] who proposed a mechanism whereby the radiation produces transitory sensitive sites on the vital molecules in the cell. The sensitizers then combine with these sites and inhibit "repair processes" that may take place during recovery. Oxygen was also believed to interfere with this combination. Lee, Anderson, and Elliker[51] demonstrated that some sensitizing compounds may also be activated by radiation and then combine with the cellular sites.

The combined use of antibiotics and radiation has been proposed with the assumption that a complementary action will be achieved. This assumption is also based on the premise that the antibiotics will have the same bacteriostatic action on the radiation survivors as on the nonirradiated bacteria. Radiation, however, destroys antibiotics at the same time that it inactivates microorganisms.[26] It is therefore important to know whether the observed complementary effect was due to a residual antibiotic activity in the irradiated food or an antibiotic effect exerted prior to, or at the time of, radiation exposure.

Data reported by Ingram and Thornley[27] suggest that the complementary effect of CTC was entirely due to the residual antibiotic activity on the radiation-surviving bacteria because the number of initially surviving microorganisms was not reduced by the presence of CTC.

The effect of sodium benzoate, potassium sorbate, and the sodium salts of methyl- or propyl-*p*-hydroxybenzoic acid suggests a similar mechanism. These agents did not affect the initial surviving numbers. They were just as effective when added after irradiation or when added prior to radiation exposure. This suggests that the presence of the above agents was not required at the time of radiation exposure, and therefore they could not have acted as radiation sensitizers. This observation also suggests that the effectiveness of these agents was not changed by irradiation. This distinction is important because it indicates that the radiation survivors are different from the nonirradiated cells to the extent that the former became susceptible to the antimicrobial agents while little effect was observed against the latter. Another important observation was that the preservatives tested were equally effective after exposure to radiation.

Several speculations can be made regarding how the microorganisms that survive irradiation became sensitive to the preservatives. One possibility is that the effectiveness of sodium benzoate is dependent on the concentration per bacterium. After irradiation most microorganisms would have been inactivated, permitting the concentration available per survivor to increase. This may have resulted in increased sensitivity among the survivors. In order to test this hypothesis, the mixed culture from the fillets was serially diluted in broth and incubated with 0.1% sodium benzoate. The decreased microbial concentration, however, did not result in increased sensitivity.[46] Another possibility is that the radiation might have eliminated those cells which are resistant to the preservatives, leaving only the sensitive microorganisms among the survivors. When approximately 40 different species from 15 genuses of microorganism were tested, some variation in tolerance to the preservatives was observed. Generally, Gram-positive cocci were most resistant and the *Bacillus* species the least. None of the cultures examined, however, have showed[49] any sensitivity to 0.1% preservative at neutral *p*H.

Our current hypothesis is that the increased sensitivity observed among the survivors toward additives is of a physiological nature and that microorganisms become susceptible to additives as a result of radiation exposure. The preservatives are believed to antagonize repair processes and prevent recovery from radiation damage.

## Feasibility of Combination Process with Chemical Agents

Radiation sensitizers will be particularly effective in sterilization processes, where increase in the lethal effect of radiation is a primary concern. Antibiotics and the chemical preservatives are best suited for low-dose food-irradiation processes, where control of postirradiation survivors is important. Regardless of the nature or the extent of benefit a food additive may bring, the acceptability of

such a process depends on proved safety. Miller[52] properly pointed out the nature of food additives by saying that there are no harmless substances but only harmless ways of using them. Even the food preservatives currently permitted by the FDA must be reexamined for combined use with radiation. The emphasis placed on the mode of action is important for this reason.

The most practical way for potentiating radiation with chemical additives is to treat the food with chemical agents prior to radiation exposure. We must therefore know the effect of radiation on the chemicals. Consideration also has to be given to the possibility of interaction between chemicals and food ingredients. Segner, Schmidt, and Boltz[45] observed that the outgrowth time of *C. botulinum* type E was doubled in the presence of 0.1% sodium benzoate in fish homogenate. They were, however, unable to demonstrate the same effect in broth, and the benzoate might have been antagonized by a constituent of the growth medium. Opposite results were reported with vitamin $K_5$, where antagonism was thought to be caused by the action of a food ingredient.[39]

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Section

IV

Isotopes and Radiation Technology

# Process Radiation Development

## Radiation-Induced Polymerization

In recent years the polymerization of various materials has been studied extensively in the plastics and related industries. A number of processes based on initiators or catalysts are being used to impart desired properties to materials used for electronics, consumer goods, and military equipment. The potential usefulness of radiation as a catalyst for the manufacture and upgrading of familiar materials and for the production of new ones is gradually being realized.

Previous issues of this review,[1,2] as well as a paper by Machurek, Dietz, and Stein,[2] have summarized commercial and near-commercial processes that use radiation in polymerization reactions. Current commercially produced materials include polyvinyl acetal resin and polyethylene wire insulation, shrinkable tubing, and plastic film. Processes soon to be "on-stream" include radiation curing of coatings, improvement of textiles, polymerization of ethylene, and production of wood–plastic combinations.[3] A number of other materials and processes are being studied under Division of Isotopes Development (DID) contracts (Table IV-1), and recent literature concerning these and other studies will be reviewed in a series of articles of which this is the first.

Recently a Russian reviewer[5] noted several reasons why radiation polymerization is one of the major practical applications of ionizing radiation:

1. It frequently takes place by a chain mechanism, resulting in high yields.
2. Under certain conditions it proceeds by an ionic mechanism, and polymers with unusually valuable properties may be prepared.
3. It takes place more smoothly than other free-radical polymerizations and can therefore be more readily and accurately controlled.
4. It produces a polymer almost as pure as the monomer, whereas in conventional methods, additives contaminate the product.
5. It can polymerize some materials that cannot be polymerized by other means.

In connection with the last advantage, a report[6] of the 1964 Netherlands meeting on large radiation sources in industry[7] stated the consensus of 150 scientists and industrialists:

> ...the key to any great leap forward in industrial uses of radiation is finding applications where the unique properties of high-energy radiations are fully exploited.... They see as the most promising fields those where high-energy radiation produces singular effects — polymerization of unsaturated hydrocarbons, modification of surface properties, grafts of protective coatings on glass and ceramics, formation of conductive layers on plastics, and the like.

A large amount of data is available on the mechanisms of radiation-induced polymerization under different conditions in liquid, solid, and gas phases. Some of the workers interested in practical applications, in addition to tapping this fund of knowledge, plead for additional theoretical studies — on the mechanism of grafting, the location of the grafted polymer, and the grafting of copolymers. In efforts to shed light on new uses, radiation has been compared to other polymerization agents (chemical catalysts, for instance), but other approaches, such as combinations of grafting and other treatments, still require detailed study to adapt specific processes to commercial application. Some of these approaches have been rewarding. As a result of studies of radiation parameters, the efficiency of radiation polymerization reportedly[8] was boosted 10-fold over previously determined values. In other work the addition

Table IV-1 DIVISION OF ISOTOPES DEVELOPMENT PROGRAMS IN RADIATION POLYMERIZATION*

| Description | Investigator | Subject of study |
|---|---|---|
| Application of $^{85}Kr$ | A. M. Feibush, Air Reduction Co. | Styrene, methylvinyl ether, chlortrifluoroethylene, vinyl acetate, acetylene, unsaturated hydrocarbons and oxygen, polyketones, and aldehydes; graft copolymerization of various monomers to rubber; adhesive curing of polymeric materials at room temperature; and reactions in gas phase |
| Radiation-induced changes in polymers and their influence on graft polymerization | F. A. Sliemers, Jr., Battelle Memorial Institute | Methacrylate and acrylate polymers; postirradiation techniques |
| Development of high-dielectric-constant, low-dielectric-loss polymer | M. Luttinger, Battelle Memorial Institute | Comparison of properties of conventional and radiation-induced polymers |
| Polymerization of ethylene | B. Manowitz, Brookhaven National Laboratory | Ethylene and formation of ethylene copolymers |
| Properties of polymers resulting from posteffect polymerization | W. H. Burrows, Georgia Institute of Technology | Monomers such as methyl methacrylate; physical properties and structural characteristics |
| Combined effects of ultrahigh pressure and radiation on organic monomers and polymers | C. J. Marsel, College of Engineering, New York University | Acrylamide, tetracyanoethylene, tetraphenylethylene, triphenylethylene, tetrabromoethylene, tetraiodoethylene, and trioxane |
| Modification of textile fiber properties by radiation-induced graft polymerization | H. A. Rutherford, North Carolina State of the University of North Carolina | Grafting of volatile vinyl monomers to fibrous polymers; polyacrylonitrile, cellulosics, cellulose esters, polyamides, and polypropylene |
| Mechanism of radiation-induced gelation in polymer–monomer mixtures | Radiation Applications, Inc. | Polyethylene, polypropylene, allyl methacrylate, allyl acrylate, divinyl benzene, diallyl maleate, polyisobutylene and cellulose acetate |
| Upgrading of by-product polypropylene by radiation techniques | Radiation Applications, Inc. | Polypropylene, polyfunctional monomers |
| Polymerization of unusual monomers | Vivian Stannett, Research Triangle Institute | Vinyl monomers and nonvinyl monomers such as cycloaliphatics, allene, nitriles, aldehydes, ketones, and ring compounds |
| Radiation-induced emulsion polymerization | Research Triangle Institute | Polymethyl methacrylate, polyisoprene, and styrene–methyl methacrylate copolymers; critical evaluation of radiation-induced emulsion polymerization |
| Reactions in swollen polymers | Zoila Reyes, Stanford Research Institute | Natural and synthetic swollen polymers; casein, vinyl monomers, methyl methacrylate, and styrene |
| Wood–plastic combinations | | |
| 1. Technical | J. A. Kent, West Virginia University | Development of process with various monomers and physical conditions |
| | J. Harry Frankfort, Vitro Engineering Division, Vitro Corp. of America | Engineering study |
| 2. Economic | Arthur D. Little, Inc. | Market potential of wood–plastic combinations |

*Adapted from Ref. 4 and other AEC sources. In this table, as in the remainder of this article, organic nomenclature is that in common use. The reader is referred to the original articles for structure and other details.

Table IV-2 IRRADIATION DOSES REQUIRED FOR TYPICAL POLYMERIZATIONS*

| Dose, Mrads | Process |
|---|---|
| 50 | Fluorocarbon polymerization, styrene bulk polymerization, ethylene polymerization (1600 psi) |
| 30 | Rubber vulcanization |
| 10 to 50 | Polyethylene cross-linking |
| 5 | Styrene emulsion polymerization, methyl methacrylate bulk polymerization |
| 2 | Acrylamide polymerization (solid state) |
| 1 | Grafting of styrene to polyethylene |
| 0.1 | Grafting of vinyl pyridine to polyethylene and of acrylonitrile to polyethylene, acrylic acid polymerization (solid state) |

*Modified from Ref. 8.

of polyfunctional monomers reduced the radiation required to cross-link polymers to as little as one-tenth the original amount; thus polymers such as polyisobutylene and polycellulose acetate, which normally are degraded when irradiated, could be successfully cross-linked.[9] Table IV-2 gives the irradiation doses required for typical polymerizations.

Workers in the general field of polymerization sometimes find unexpected parallels between their research and that of the life scientists. These fundamental insights probably will open up still other areas of application. An authority[10] recently said:

> The nature of the last unit in the growing chain and specific sequences of units attached to it have important effects on the addition of either monomer. In genetics, growth, and healing, similar mechanisms are involved. The processes by which the brain stores and uses information have parallels with the copolymerization process.
>
> ...It seems that the most startling advances will be in the area of synthetic polymers and copolymers for biological use...drugs, agents to promote healing processes, or as aids to other important physiological processes.

## Background

The explosive growth of the field of polymerization was triggered in part by the lack of natural rubber in the early 1940's. Physical and chemical stimuli were used to solve this problem, and a most significant result of wartime research in catalytic polymerization was styrene–butadiene rubber. Even at that time, however, work in radiation-induced polymerization was going on; in fact, study of the action of ionizing radiation on compounds followed soon after the discovery of X rays* and was intensified[13] in the 1920's. In 1944 Rexer noted accelerated polymerization with gamma and X rays,[14] and three years later the effect of these radiations on acrylonitrile in aqueous solution was reported.[15] One of the first peacetime irradiations in the Clinton Pile involved plastics.[16] According to a literature survey,[17] the first reference to radiation-induced solid-state polymerization was by Schmitz and Lawton[18] in 1951, who noted that tetraethylene glycol dimethacrylate polymerized explosively after irradiation of the solid monomer. In 1952 a paper by Charlesby[19] on the cross-linking of polyethylene stimulated the whole field of high polymer radiation physics.[20] By 1956 Restaino et al.[21] had presented results on the polymerization of acrylamide, a monomer that has received much attention in the literature.†

*REVIEWS AND SURVEYS*

A number of reviews and bibliographies on polymerization have been published. Some of these deal specifically with radiation, such as Charlesby's book.[22] Others, like Güterbock's 1959 literature and patent survey of polyisobutylene and isobutylene copolymerizates,[23] appear in the general polymer literature.‡ Kennedy[24] gives a table summarizing all available references to monomer pairs published in the literature up to early 1962. Reactivity ratios are given only for isobutene–styrene and styrene–methyl acrylate in studies done with gamma radiation. Kennedy, who concludes that polymerization initiated with high-energy radiation probably proceeds by cationic mechanisms, especially at low temperatures, points out a number of studies in the field of cationic copolymerization with gamma rays to initiate the reaction.[25]

Polymerization of a monomer in the solid state by conventional catalysis is difficult. In the solid-state survey previously mentioned,[17] more than 20 monomers are reported to polymerize by radiation, but these reviewers conclude:

> Perhaps the most relevant feature of the studies on solid state polymerization has been the difficulty encountered by the various authors in positively assigning any one mechanism to the reaction. It is likely that in some cases both free radical and ionic mechanisms operate in the solid state.
>
> The rates of polymerizations in the solid state... are at maximum near to the monomer melting point but they are generally low when compared with liquid or solution polymerizations. Some monomers, however, will only polymerize using ionizing radiation in the solid state (e.g., cyclic monomers).... The presence of water has been found to accelerate some of the polymerizations....

Extensive tables of physical conditions and yields are given in Ref. 17; an abridged version of the monomer data is shown in Table IV-3. Particular attention in their review centers on the relatively few monomers that yield stereospecific polymers in the solid state. As a group the cyclic monomers (Table IV-4) yield the best-oriented and most-crystalline polymers. Evaluation of the physical and mechanical properties of the polymer from 3,3-*bis*(chloromethyl)-oxacyclobutane (Penton) suggests that the polymers from cyclic monomers might be useful for the production of oriented films and for injection moldings. The latter

*According to a recent textbook,[11] work on radiation-initiated polymerization antedates the discovery of X rays. The Thenards[12] in 1874 described the formation of an inert solid when acetylene was exposed to an electric discharge.

†The polymerization of acrylamide (m.p. 84°C) and related monomers has been studied over a wide range of temperatures (– 196 to 65°C) and dose rates ($3 \times 10^3$ to $9 \times 10^5$ rads/hr). G-initiation values of about 0.5 to 0.8, a relatively low value, have been calculated.

‡For lower molecular weight isobutene copolymerizates, consult Ya. T. Eidus and B. K. Nefedov, Polymerization of Isobutylene, *Russian Chemical Reviews (English Translation)*, 29(7): 390-410 (July 1960).

Table IV-3 POLYMERIZATION OF SELECTED MONOMERS IN THE SOLID STATE*

| Monomer | Melting point, °C | Polymerization | Post-polymerization | Polymerization temp., °C | Dose rate, rads/hr | Polymerization rate, %/hr |
|---|---|---|---|---|---|---|
| Acrylamide | 84 | + | + | 30 | $1.9 \times 10^5$ | 12.8 |
| Methacrylamide | 110 | + | ? | 30 | $1.9 \times 10^5$ | 0.7 |
| *N,N*-methylene-*bis*-acrylamide | 185 | + | ? | 30 | $1.9 \times 10^5$ | 1.6 |
| Acrylic acid | 11 | + | ? | −18 | $3.1 \times 10^5$ | >400 |
| Methacrylic acid | 16 | + | ? | −18 | $3.1 \times 10^5$ | >400 |
| Calcium acrylate | >100 | + | + | 35 | $1.9 \times 10^5$ | 18.1 |
| Potassium acrylate | >100 | + | + | 35 | $1.9 \times 10^5$ | 56.8 |
| Barium acrylate | >100 | + | + | 35 | $1.9 \times 10^5$ | 75.1 |
| Vinyl stearate | 34 | + | ? | 26 | $1.9 \times 10^5$ | 20.0 |
| Vinyl carbazole | 65 | + | ? | 30 | $2.2 \times 10^5$ | 1.1 |
| Acrylonitrile | −83 | + | − | −196 | $3.8 \times 10^4$ | 0.8 |
| Methacrylonitrile | −40 | + | + | −78 | $3.2 \times 10^5$ | 1.0 |
| Butadiene | −109 | + | ? | | | |
| Styrene | −30 | + | + | −78 | $2.1 \times 10^6$ | 0.17 |
| Methyl acrylate | −75 | − | − | | | |
| Methyl methacrylate | −50 | − | + | | | |
| Formaldehyde | −118 | + | + | −196 | $7.8 \times 10^2$ | 40 |
| Ethylenesulfonamide | 24 | + | + | 20 | $2.5 \times 10^5$ | 75 |
| Hexamethylene-cyclotrisiloxane | 64 | + | ? | 60 | $1.3 \times 10^7$ | 10 |
| Isobutene | −141 | + | ? | | | |

*Modified from Ref. 17. With the exception of hexamethylene-cyclotrisiloxane, which was irradiated in air, and isobutene, for which irradiation conditions were not stated, monomers were irradiated in vacuum.

application is discussed in several industrial journals[26] and by Sandiford.[27]

An indication of the expanding technology of the radiation chemistry of polymers is given in a partial survey of the patent literature.[28] British and U. S. patents cover polymerization by high-energy electrons,[29] and a French patent[30] covers the polymerization of butadiene in chlorinated solvents at temperatures below −40°C.

About a third of the 90 references in a bibliography by Ballantine and Kopp[31] and 22 papers at a 1962 international symposium[32] deal with radiation-induced polymerization and graft polymerization. Cross-linking and degradation reactions are stressed in a recent review of radiation effects on polymers.[33] The primary emphasis in the latter work, however, was on plastics, elastomers, resins, and components such as tires and gaskets. Information that has resulted from the use of radiation from isotopes for addition polymerization of unsaturated monomers has been summarized by Ayrey,[34] who emphasized the extensive use of tracers to study the mechanism of polymerization. A fairly comprehensive overview of the early literature can be obtained from a 1959 bibliography by Rana.[35]

In an effort to summarize and evaluate the effect of radiation on cargoes in nuclear-powered tankers and the applications for petroleum and its products in nuclear-propelled ships, Baxter and Black[13] in 1959 surveyed more than 400 unclassified reports and articles and 250 classified reports. Primary emphasis of this study was on radiation chemistry of hydrocarbons in the condensed or liquid states. In these systems chemical changes can be at-

Table IV-4 POLYMERIZATION OF SELECTED CYCLIC MONOMERS IN THE SOLID STATE*

| Monomer | Melting point, °C | Polymerization | Post-polymerization | Polymerization temp., °C | Total dose, rads | Polymer yield, % |
|---|---|---|---|---|---|---|
| Trioxane | 64 | + | + | 35 | $4.4 \times 10^5$ | 19.8 |
| Diketene | −6.5 | + | + | −78 | $1.1 \times 10^5$ | 5.29 |
| β-propiolactone | −33 | + | + | −78 | $1.4 \times 10^5$ | 13.2 |
| 3,3-*bis*(chloromethyl)-oxacyclobutane | 18.7 | + | + | 11.6 | $1.7 \times 10^4$ | 13.4 |

*Modified from Ref. 17. Irradiations were carried out in air, and fine, oriented crystalline polymer structures were obtained. Ionic mechanisms were proposed for the polymerizations.

Table IV-5 G VALUES FOR RADIATION CHANGES IN HYDROCARBONS*

| Result of radiation | G values | | |
|---|---|---|---|
| | Saturated | Unsaturated | Aromatic |
| Polymerization | None | 10 to 2000 | None |
| Cross-linking | ~1 | 6 to 14 | ~1 |
| $H_2$ evolution | 2 to 6 | 1 | 0.04 to 0.4 |
| $CH_4$ evolution | 0.06 to 1 | 0.1 to 0.4 | 0.001 to 0.08 |
| Destruction of irradiated material | 4 to 9 | 6 to 2000 | ~1 |

*Modified from Refs. 13 and 36. The G value is commonly defined as the number of molecules that react for each 100 ev absorbed in the system.

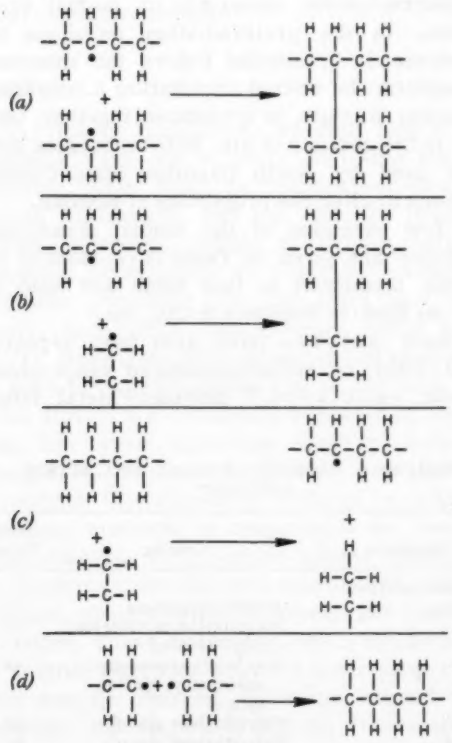

*Fig. IV-1 A few of the pertinent radical processes that occur during polymer irradiation.* (a) *Recombination of two type 1 radicals (alkyl free radicals) to form a cross-link.* (b) *Recombination of a type 1 and a type 2 radical (type 2 generated by carbon–carbon bond breakage) in an "end-linking" process to form a polymer chain branch.* (c) *Chain transfer, which can also involve type 1 radicals.* (d) *Recombination of type 2 radicals to regenerate a molecule like the parent molecule (dot indicates unpaired electron of a free radical). All the radical processes occurring in polyethylene are not represented, and the ones shown may not occur to an equal extent. The purpose of this figure is to illustrate, in simple fashion, mechanisms assumed to occur in at least some polymers.* [*From G. Adler,* Science, *141: 323 (1963).*]

tributed principally to the formation of free excited states and free radicals. Table IV-5 summarizes some of the G values for these changes. Adler[37] discusses the production of free radicals and the mechanism of cross-linking (Figs. IV-1 and IV-2).

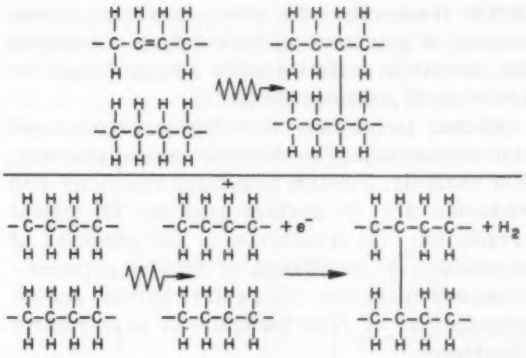

*Fig. IV-2 Some addition mechanisms that have been invoked to explain cross-linking; $e^-$ represents an electron, and the wavy arrow indicates radiation.* [*From G. Alder,* Science, *141: 323 (1963).*]

In another review[38] on the properties of ionizing radiation in experimental and industrial chemistry and on radiochemical preparation of high polymers, effects of catalysts and of radiation are compared. Vinyl chloride is emphasized in the last seven chapters of a 1963 Romanian review[39] of chemical reactions induced by ionizing radiation, and the significance of high-energy radiation effects in polymer chemistry (including effects on proteins) is discussed by Whitfield and Wasley.[20]

Shultz,[40] who reviewed the effects of ionizing radiation on synthetic polymers, called for further work on the mechanisms of chain cleavage and cross-linking, particularly in quantitative measurements of the intermediate and ultimate chemical species formed. Among the effects of radiation on polymers, cleavage and chemical bonding produce, as in most chemical reactions, the most obvious changes. The relative importance of ionic and free-radical reactions that occur during irradiation continues to be an open question. Most investigators have proposed reaction mechanisms involving free-radical species, although ionic and ion–molecular paths have been demonstrated, especially in the solid state.

## Methods of Polymerizing Materials by Radiation

In the simplest radiation polymerization system, only one component is irradiated and the amount of product formed is measured. Early work centered on effects of radiation in such simple systems. Damage to the irradiated material frequently was noted. However, cross-linking of polymers by high-energy irradiation has received more scientific attention than has cleavage of polymer chains.[41]

Recent techniques in radiation polymerization involve binary or multicomponent systems. For example, possible branching reactions with radiation are (1) surface grafting, (2) mutual irradiation, (3) irradiation in the presence of monomer, (4) irradiation of swollen polymer–monomer mixtures, (5) preirradiation, and (6) introduction of free radicals to a polymeric "backbone."

By use of surface reactions, fibers have been modified to improve the textiles made from them in regard to texture, washability, dye retention, antistatic properties, and resistance to abrasion. Film such as polyethylene has been modified to improve scuff resistance, printability, permeability to liquids and vapors, solvent resistance, and adhesion. Not all these modifications have reached practical application with the use of radiation; chemicals, heat, and other conventional treatments are more widely used at present. However, several of these processes lend themselves to radiation polymerization. Of 80 patents cited in a recent review[42] of surface reactions, about 30 involved irradiation.

Use of reactions at the surface of a solid substrate is becoming increasingly widespread. Angier,[42] of Ethicon, Inc., in discussing these "topographical reactions," gives G values for a number of monomers (Table IV-6). The primary requirement for surface grafting, according to this worker, is low solubility of the monomer in the base polymer. A number of the systems, shown in Table IV-7, exemplify the effect of radiation grafting when the penetration of the monomer into the substrate is negligible. Diffusion, oxygen, preirradiation treatment, crystallinity, and thickness of the grafted layer have considerable effect, in addition to the G value.

Likewise, a number of recent patents deal with surface modification of polymeric substrates by preirradiation in the presence of nonpolymerizable materials or mutual irradiation. In the preirradiation technique the substrate is irradiated before the monomer is applied. In mutual irradiation a polymer–monomer mixture is irradiated together, usually in the absence of air. Both techniques have been used by North Carolina State College workers to alter the properties of textiles.

A few examples of the mutual irradiation technique are given in Table IV-7. Most of the patents mentioned in this table are held by E. I. du Pont de Nemours & Co., Inc.

Recent patents have also been reported (1963–1964) on radiation-induced vapor-phase organic reactions,[43] polymer–metal lami-

Table IV-6 G VALUES OF VARIOUS MATERIALS*

| Monomer | G value |
|---|---|
| Styrene | 1.6 |
| Acrylonitrile | 2.7 |
| Methyl acrylate | 23.5 |
| Methyl methacrylate | 27.5 |
| Vinyl acetate | 33.0 |
| Isobutene (polyethylene) | 5 to 6 |
| *N*-Vinylpyrrolidone | 15.0 |

*Modified from Ref. 42, p. 1011. It is assumed that the G value of the polymer is the same as that of the monomer.

Table IV-7 PATENTED MUTUAL IRRADIATION SYSTEMS*

| Substrate | Coating | Dose, Mrads |
|---|---|---|
| Polymer–polymer | | |
| Nylon | Polychlorostyrene | 40 |
| | Poly(vinylidene chloride/vinyl acetate) | 40 |
| | Poly(methoxynonaethyleneoxyethyl methacrylate) | 40 |
| | Poly(ethylene glycol) | 40 |
| Wool | Poly(ethylene glycol) | 40 |
| Silk | Poly(ethylene glycol) | 40 |
| Rubber | Poly(ethylene glycol) | 40 |
| Polytetrafluoroethylene | Poly(ethylene glycol) | 27 |
| Poly(methyl methacrylate) | Poly(allyl methacrylate) | 20 |
| Poly(styrene-*co*-butadiene) | Poly(oxyethylene glycol) | 10 |
| Polyethylene | Poly(ethylene glycol) | |
| Polymer–nonmonomer | | |
| Nylon | Hexamethylenediamine | † |
| Polyacrylonitrile | Dodecafluoroheptyl alcohol | † |
| Nylon | Palmitic acid | ‡ |
| Polyethylene | Glycerol | ‡ |

*Modified from Ref. 42.
†No dose given; 2-Mev electrons used.
‡No dose given; alpha particles used.

nates,[44] treating of cotton with radiation,[45] and curing of magnetic tape.[46]

New methods of using radiation polymerization as a tool are continually being developed. Manno[47] recently suggested the use of radiation to screen large numbers of monomers under polymerization conditions so that a rapid survey of various combinations could be made to find promising polymers. By this program, two fluoropolymers—polyvinyl fluoride and the copolymer of chlorotrifluoroethylene and ethylene—were selected. A commercial radiation process for producing the latter copolymer apparently could be competitive with conventional catalytic methods.

## Textiles

All fibrous polymers, whether natural or synthetic, lack one or more desirable market qualities when used in textiles—dyeability, crease resistance, and the like. To improve natural polymers, textile manufacturers permanently modify them by acids and catalysts. Synthetic polymers for the most part melt easily, dye poorly, and develop static electricity. Relatively little research has been done on ways to improve either type of polymer.

All fibers are ultimately degraded by radiation, but some materials undergo desirable changes at intermediate points. Cross-linking, a reaction that cannot be produced by ordinary chemical methods, is responsible for many of these changes. Many simple organic molecules, particularly those that are unsaturated, undergo changes rapidly with relatively low doses of radiation. These molecules, which can be added to textile fibers to improve their properties, thus can be changed while the natural fiber remains undamaged. Modifying the relatively unstable cellulosic fibers by this method is attractive since these comprise nearly 90% of the total fiber consumption in the United States. Because they are relatively stable, synthetic polymers perhaps could be modified after they are in fibrous form; radiation exposure in media that will promote cross-linking or grafting is a promising technique.

Armstrong et al.[48–53] made a broad study of methods for modifying textiles at North Carolina State of the University of North Carolina (NCS) at Raleigh, N. C., from 1958 to 1963 under a DID contract and with some funds from interested textile firms: Burlington Industries, Inc., Cone Mills, Inc., Reeves Bros., Inc., and J. P. Stevens & Co., Inc.

Although they ruled out the method of irradiation alone, NCS workers demonstrated that graft polymerization of fibers by irradiation in the presence of monomers in the vapor phase is a promising way to improve textile properties. Because they obtained uniform results with this technique, they recommended it as the method of choice for commercial application.

Various textile fibers were irradiated at NCS in Gammacell-220 $^{60}$Co facility (Fig. IV-3). The apparatus, physical conditions, and flow rates were varied for different experiments.

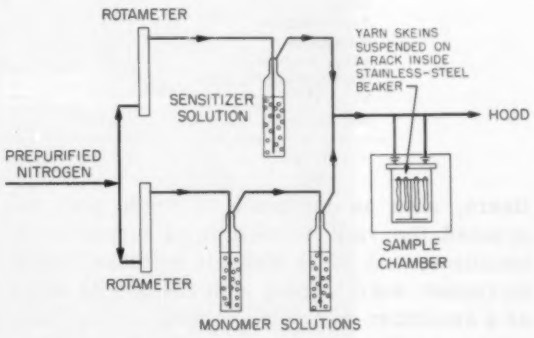


*Fig. IV-3 Apparatus for vapor-phase graft polymerization of textile fibers.*

The experimental procedure for a general survey of grafting various vinyl monomers to textile fibers was:

1. One-gram skeins of each yarn were suspended on a rack in a stainless-steel beaker in the Gammacell sample chamber.
2. The beaker was flushed for 2 hr with prepurified nitrogen saturated with monomer vapor.
3. The beaker was lowered to the irradiation position, and flushing was continued during a predetermined irradiation period.
4. The monomer addition was determined from the weight gain of the yarn sample.

With two fibers, acetate and polypropylene, methyl acrylate was added in significant quantity. Six other monomers were added in various amounts to these fibers, but neither these nor methyl acrylate was added significantly to rayon, cotton, wool, polyester, or acrylic yarns. Acrylic acid addition to nylon was 2.4%, the

best percentage for the various monomers tried with this fiber.

Higher addition of monomers to acetate, polypropylene, and nylon and limited addition to cotton, rayon, and wool were obtained by a modified procedure with the use of sensitizers, which magnified the effect of radiation. Table IV-8 shows some of the results obtained with water as sensitizer. Most of the other sensitizers, such as methanol or acetic acid, increased the rate of addition of acrylonitrile, butadiene, and vinyl chloride to these fibers. Increases were largest with the use of water as a sensitizer on cotton and wool.

Table IV-8 ADDITION OF MONOMER IN THE PRESENCE OF WATER IN VAPOR-PHASE RADIATION GRAFT POLYMERIZATION

| Monomer | Monomer added, % | | | | | |
|---|---|---|---|---|---|---|
| | Acetate | Polypropylene | Nylon | Cotton | Rayon | Wool |
| Methyl acrylate | 72.3 | 51.8 | 12.6 | 19.2 | 2.2 | 1.8 |
| Ethyl acrylate | 106.0 | 23.3 | 7.2 | 0.9 | 1.0 | 1.4 |
| Acrylonitrile | 30.8 | 4.3 | 5.6 | 2.1 | 3.2 | 4.4 |
| Butadiene | 24.0 | 5.7 | 2.8 | 0.0 | 0.0 | 0.3 |
| Vinyl acetate | 14.6 | 4.7 | 1.5 | 0.0 | 0.1 | 0.0 |
| Methyl methacrylate | 18.2 | 2.4 | 0.8 | 0.5 | 0.3 | 0.1 |
| Vinyl chloride | 3.0 | 2.9 | 2.0 | 0.0 | 0.0 | 0.1 |
| Styrene | 0.5 | 1.1 | 0.2 | 0.0 | 0.0 | 0.0 |

Water, although necessary for polymerization, was not required *during* irradiation. It was found that monomer could be added *after* irradiation if water vapor were present. Some results from postirradiation graft polymerization are given in Table IV-9.

A study was made of the effects of several variables on the rate of grafting so that a satisfactory process could be designed. The vapor-phase process was diffusion controlled; therefore an increase in carrier-gas flow rate increased the rate of grafting. Oxygen inhibited the polymerization. An increase in temperature usually decreased the rate of grafting (for a constant carrier-gas–monomer concentration) because the amount of monomer sorbed decreased with an increase in temperature.

The rate of grafting depended on the fiber and monomer. In general, the order of reactivity of the fibers was acetate > polypropylene > nylon > cotton > rayon > wool > polyester > acrylic. The order of reactivity of the monomers studied was methyl acrylate > ethyl acrylate > acrylonitrile > butadiene > vinyl acetate > methyl methacrylate > vinyl chloride > styrene. In the case of grafting of acrylonitrile to cotton, the rate was proportional to the 0.4 power of the acrylonitrile content and to the 2.4 power of the water content of the carrier gas.

The rate of grafting to cotton was increased by chemical modification of the cellulose. Partial acetylation, hydroxyethylation, or cyanoethylation of cotton increased the rate of grafting of acrylonitrile to cotton.

Table IV-9 POSTIRRADIATION VAPOR-PHASE GRAFT POLYMERIZATION (With and Without Water)

| Monomer | Monomer added, % | | | | | |
|---|---|---|---|---|---|---|
| | Acetate | Polypropylene | Nylon | Cotton | Rayon | Wool |
| Methyl acrylate | 32.3 | 4.0 | 0 | 0 | 0 | 0 |
| With $H_2O$ | 58.8 | 10.9 | 1.8 | 3.3 | 1.1 | 0.8 |
| Ethyl acrylate | 0.8 | 1.4 | 0.1 | 0 | 0 | 0 |
| With $H_2O$ | 31.7 | 4.0 | 0.8 | 0.2 | 0.3 | 0.3 |
| Acrylonitrile | 11.3 | 0.9 | 0.7 | 0 | 0 | 0 |
| With $H_2O$ | 7.3 | 0.9 | 2.5 | 0.2 | 0.6 | 1.3 |
| Butadiene with $H_2O$ | 7.5 | 5.0 | 0.9 | 0.3 | 0 | 0 |
| Vinyl acetate | 3.6 | 0.3 | 0.1 | 0.2 | 0.2 | 0.2 |
| With $H_2O$ | 1.6 | 0.2 | 0.6 | 0 | 0.1 | 0.3 |
| Methyl methacrylate | 0 | 0.7 | 0 | 0 | 0 | 0 |
| With $H_2O$ | 5.4 | 0.7 | 0.2 | 0 | 0 | 0 |

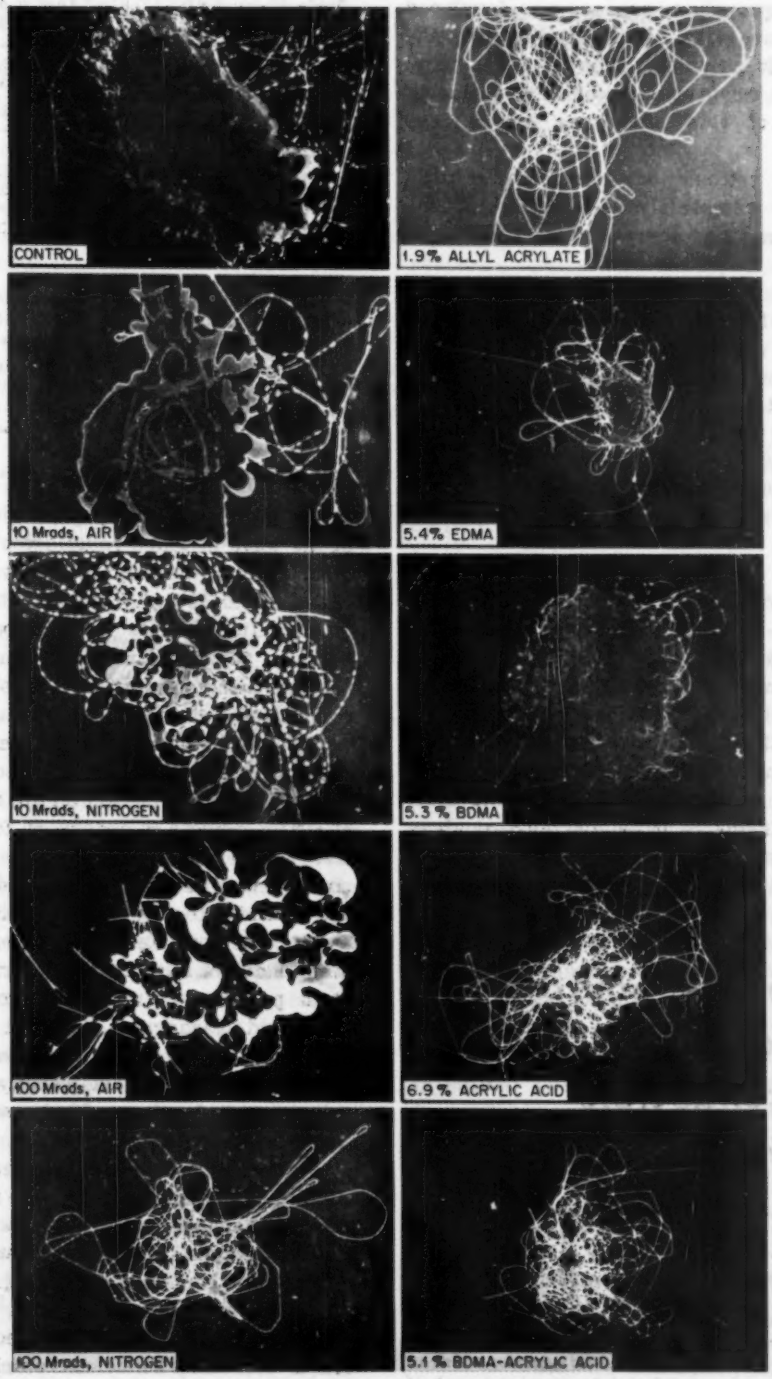


*Fig. IV-4 Effect of heating nylon to 290°C after treatment with radiation alone (left) and after radiation cross-linking with monomers (right). All pictures are shadowgraphs. Monomers were irradiated at 1 Mrad in nitrogen (500 cm³ of nitrogen bubbled through methanol, 500 cm³ through monomer) with the monomer in the vapor phase. EDMA, ethylene dimethacrylate; BDMA, butylene dimethacrylate.*

After the vapor-phase process was developed to a satisfactory point, it was used to study the effect of graft polymerization of some vinyl monomers on the properties of various fibers. The results of this study are summarized below:

1. Grafting cotton with acrylonitrile produced a fiber completely resistant to microorganism attack and increased its resistance to heat.

2. Grafting polypropylene fiber with vinyl acetate, methyl acrylate, or ethyl acrylate gave a product that was dyeable with dispersed dyes.

3. Grafting nylon and polypropylene with styrene improved weather resistance.

4. Grafting cotton cloth with allyl acrylate followed by cross-linking by radiation and grafting with methylol acrylamide and chemical curing improved crease-recovery properties (Table IV-10).

5. Grafting nylon yarn with allyl acrylate, acrylic acid, or a combination of acrylic acid with a cross-linking monomer increased the dimensional stability at 290°C (Fig. IV-4). Increases in modulus of elasticity of 30 to 50% were obtained by grafting with acrylic acid alone or in combination with a cross-linking monomer.

6. Irradiating fibers alone did not improve their physical properties. Solubility data indicated cross-linking of nylon and polypropylene at 100 Mrads in nitrogen and of an acrylic fiber at 10, 50, and 100 Mrads in either nitrogen or air. (D. A. Fuccillo)

Table IV-10 WRINKLE RESISTANCE AND STRENGTH OF GRAFT-POLYMERIZED AND UNTREATED COTTON

| Sample | Recovery angle,* deg | | Tear strength, lb |
|---|---|---|---|
| | Dry | Wet | |
| 9% allyl acrylate grafted to cotton at 10 Mrads | 247 | 273 | 0.9 |
| Control | 180 | 200 | 1.4 |

*An acceptable recovery angle value is 240°.

# Radioisotopic Sources To Study Radiation Damage in Semiconductors

By J. W. Cleland*

*The ways in which radioisotopic sources have been used in the Solid State Division of the Oak Ridge National Laboratory for the study of radiation-induced lattice defects in semiconductors are described, and additional uses that may become important with advances in radioisotope production and source development are presented. Two types of sources have been used: (1) gamma sources and (2) neutron sources in which neutrons are produced by bombardment of beryllium with high-energy photons or alpha particles. Agreement of calculated with observed defect-introduction rates improved with increase in photon energies. Neutron fluxes from $^{124}Sb$–Be (50 curies), $^{210}Po$–Be (90 curies), and $^{239}Pu$–Be (10 curies) were too low for most radiation-damage experiments; however, $^{241}Am$, $^{244}Cm$, and $^{252}Cf$ should be useful in future studies. Irradiation of n type germanium and silicon with $^{241}Am$–Be neutron sources gave conduction-electron removal rates of approximately 13.4 and 11.5 per incident neutron, respectively.*

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The purpose of this paper is to indicate some of the ways in which radioisotopic sources have been used in the Solid State Division of the Oak Ridge National Laboratory (ORNL) for a study of radiation effects in semiconductors and to suggest additional uses that may become important with continued advances in radioisotope production and source development.

The mechanism by which energetic radiation introduces defects in solids has been described.[54] Information on the type and number of lattice defects is necessary for any quantitative test of radiation-damage theory and for construction of a realistic model of the nature and distribution of radiation-induced defects. Two classes of defects have been distinguished: simple defect structures comprising the relaxation products of radiation-produced interstitials and vacancies (Frenkel type defects), designated type *a*; and disordered regions or regions of high defect density whose spatial extent may be from 50 to several hundred angstroms, designated type *b*.

Monoenergetic-electron irradiation experiments have established that the minimum recoil energy transferred to a lattice atom for its displacement to a stable interstitial position is about 15 ev for semiconducting materials[55] and about 25 ev for most metals.[54] These values correspond to a threshold electron energy of 0.3 to 0.7 Mev; hence extremely thin samples were required for the experiments. The problems associated with depth of penetration can be avoided, and the equivalent of internal electron bombardment can be attained by use of gamma rays. The primary mechanism for the

transfer of photon energy (0.5 to 3.0 Mev) from a radioisotopic source to a lattice atom is an indirect interaction involving (1) photoelectron production, (2) pair production (electron–positron), and (3) Compton-electron production. Since the absorption coefficient of photons is orders of magnitude less than that of electrons in this energy range, radioisotopic photon sources can produce type *a* defects in bulk specimens without the experimental problems associated with thin films.

Fast neutrons and heavy charged particles have been shown to produce a mixture of type *a* and type *b* defects. An important experimental quantity is the minimum bombarding-particle energy or lattice-atom recoil energy required to produce a defect cluster or disordered region.[56] The obvious way to determine this value is to irradiate specimens with monoenergetic neutrons to ascertain the minimum energy required to cause a change in a property known to be sensitive to disordered regions. Therefore photoneutron sources that produce essentially monoenergetic neutrons whose energy depends on the gamma energy and the binding energy of the target nucleus are valuable research tools for this purpose. Also of interest is the extent of lattice damage as a function of neutron energy for relatively high-energy (>1 Mev) neutrons. Information of this type is needed in the apportioning of the primary recoil energy between ionization losses and elastic-collision losses. The average fission-neutron or pulsed-reactor spectrum has a peak neutron energy of about 1 Mev, and the spectrum is a sensitive function of both the type of reactor and the position within the reactor. The neutron output of an $(\alpha,n)$ source, on the other hand, may have a peak neutron energy of 4 to 5 Mev. Therefore sources of this type are also of considerable interest in radiation-damage studies.

## Radioisotopic Sources

The availability of large quantities of reactor-produced radioisotopes of high specific activity has led to the construction of many irradiation facilities that contain kilocurie or even megacurie amounts of radioactive material. Technical bulletins and catalogs are available[57] which describe photon, alpha, beta, heat, and neutron sources, and a survey[58] was made recently of the experimental facilities for 88 different gamma irradiators. Table IV-11 lists some of the radioisotopic sources that have been used in the ORNL Solid State Division for neutron and photon studies of radiation effects in solids. The photon intensity was determined in most cases by the ceric–cerous dosimetry technique. The photon flux was calculated from the relation

$$\phi = \frac{R}{EK\mu}$$

where $\phi$ = photon flux per square centimeter per second
$R$ = source strength in roentgens per hour determined by dosimetry
$E$ = photon energy
$K$ = constant for conversion of units
$\mu$ = absorption coefficient, assumed to be $3.5 \times 10^{-5}$ for photon energies of 0.5 to 3.0 Mev

Table IV-11 NEUTRON AND PHOTON SOURCES USED AT ORNL FOR STUDY OF RADIATION EFFECTS IN SOLIDS

| Source material | Source strength, curies | Photon energy, Mev | Half-life | Source strength, r/hr | Flux,* photons/(cm²)(sec) (source surface) | Be $(\gamma,n)$ flux, n/(cm²)(sec) (source surface) | Neutron energy, Mev |
|---|---|---|---|---|---|---|---|
| $^{137}Cs-^{137m}Ba$ | 9,000 | 0.667 | 30 years | $1.9 \times 10^6$ | $1.55 \times 10^{12}$ | - | - |
| $^{60}Co$ | 11,000 | 1.17 and 1.33 | 5.3 years | $4.9 \times 10^6$ | $2.10 \times 10^{12}$ | - | - |
| $^{124}Sb$ | 1,000 | 1.7 (70%) | 60 days | $1.4 \times 10^5$ | $4.30 \times 10^{10}$ | $4.3 \times 10^6$ | 0.035 |
| $^{24}NaF$ | 280 | 2.76 (50%) | 15 hr | $1.1 \times 10^5$ | $1.4 \times 10^{10}$ | $1.4 \times 10^6$ | 0.82 |
| $^{24}NaF$ (SRE)† | ‡ | 2.76 (50%) | 15 hr | $1.3 \times 10^5$ | $1.7 \times 10^{10}$ | $1.7 \times 10^6$ | 0.82 |
| Cd gammas | - | >3.0 (25%) | - | - | 70% $n\upsilon_{th}$ | $10^{-4}$ $n\upsilon_{th}$ | Spectrum |
| $^{140}Ba-^{140}La-^{140}Ce$ | - | 2.5 (0.004%) | - | $4.5 \times 10^4$ | - | $\sim 10^6$ | 0.62 |

*$n\upsilon_{th}$ = thermal-neutron flux.
†Sodium Reactor Experiment (SRE).
‡0.28 curie per cubic centimeter of NaF coolant in the intermediate heat exchanger (SRE).

The beryllium $(\gamma,n)$ cross section is only about $10^{-3}$ barn for photons in this energy range; hence the expected neutron flux is only about one ten-thousandth that of the measured photon flux for these sources. The calculated neutron flux (source surface) and the neutron energy for the beryllium $(\gamma,n)$ reaction are also indicated in Table IV-11.

The first $^{60}$Co gamma-source facility for the Solid State Division was constructed in 1956 and is similar to that described by Burton, Ghormley, and Hochanadel[59] except that the entire assembly terminated at floor level. This design was chosen because it used the standard ORNL radioisotope shipping container. Subsequent facilities, which are identical with the exception of water cooling, have also been constructed, and the shipping container has been used to transfer and reload both $^{60}$Co and $^{137}$Cs source tubes that contained as much as 11,500 curies. Twelve additional sample tubes external to the source tubes in the $^{60}$Co and $^{137}$Cs facilities have a photon flux about one-third that listed in Table IV-11 for the central exposure positions.

The antimony–beryllium source was devised by Hennelly.[60] His facility, composed of five sources that have a total output of about $10^{10}$ n/sec from 1000 curies of antimony, was originally designed to supply neutrons for a pressurized subcritical experiment for reactor-physics studies of heavy-water-moderated power reactors. The antimony is replaced four times a year to maintain this convenient source of 1.7-Mev photons, 0.025-Mev neutrons, and, with a moderator, thermal neutrons.

Two sources of photons and neutrons containing $^{24}$Na have been used: (1) An isotope capsule was filled with 11.2 g of fused NaF and irradiated 24 hr in the Oak Ridge Research Reactor (ORR) at an estimated thermal-neutron flux of $2 \times 10^{14}$ n/(cm$^2$)(sec). This source at its surface provided a total dose of $1.5 \times 10^{15}$ (2.76 and 1.38 Mev) photons/cm$^2$ in the first 24 hr of decay. (2) Molten alkali fluoride salt is used as a heat-exchange medium in certain loop experiments, test reactors, and research or power reactors. The saturation activity of the sodium in the sodium fluoride coolant in the intermediate heat exchanger of the North American Aviation (Atomics International) Sodium Reactor Experiment (SRE) is 0.28 curie/cm$^3$, and, at a reactor power level of 10 Mw, dosimeter experiments have indicated that the photon intensity inside the heavily shielded room at the surface of the intermediate heat exchanger is $1.7 \times 10^{10}$ photons/(cm$^2$)(sec), as shown in Table IV-11. It is anticipated that an extremely powerful source of 2.76- and 1.38-Mev photons from $^{24}$Na, 0.82-Mev neutrons from beryllium, and 0.2-Mev neutrons from $D_2O$ can be obtained from the SRE when the power level is increased to 30 Mw in 1965.

In addition to static radioisotopic photon and neutron sources, the $(n,\gamma)$ reaction in a nuclear reactor can serve as a valuable source of energetic photons and neutrons. When cadmium metal is placed in a thermal-neutron flux, capture gamma rays are emitted, and these have been used to produce radiation damage in semiconducting materials.[61] It has been suggested that these gammas might also be used to produce photoneutrons from $D_2O$ or beryllium.

An alternative dynamic approach was suggested by Almqvist,[62] who examined the $^6$Li$(n,\alpha)^3$H reaction in a thermal-neutron flux and then the subsequent $^6$Li$(t,n)^8$Be reaction, which produced $2.7 \times 10^{-4}$ (15.3 Mev) neutron per triton. However, there is a fundamental limitation that should be considered in all radiation-effects experiments that concern neutron production in the thermal-neutron region of a reactor: the fast-neutron (damaging) background flux from the reactor itself can be estimated from the cadmium ratio, and this flux must be less than that available from the $D_2O$, beryllium, or the $^6$Li$(t,n)$ reaction if the fluxes from these sources are to be studied specifically. The available flux from such sources is only about one ten-thousandth that of the thermal neutrons; hence one must have a facility with a large cadmium ratio to minimize the effect of any background of fast (damaging) neutrons.

Spent fuel elements have also been used as a source of gamma rays for photoneutron reactions in irradiation experiments. One of the prominent fission products in a reactor fuel is $^{140}$Ba (12.8-day half-life), which decays to $^{140}$La (40-hr half-life). It has been estimated that the $^{140}$La $\rightarrow$ $^{140}$Ce(stable) reaction accounts for about 6% of the total photon activity from a reactor fuel element. A few percent of the $^{140}$La $\rightarrow$ $^{140}$Ce decay is that of a 2.5-Mev gamma, which would produce a 0.62-Mev photoneutron from beryllium.

Preliminary experiments were conducted in the gamma grid in the pool of the ORR, and the

total activity at the center was determined to be $1.25 \times 10^7$ r/hr when the grid was loaded with four ORR fuel elements. This value corresponds to $7.5 \times 10^5$ r/hr for the $^{140}$La, of which $4.5 \times 10^4$ r/hr is from the 2.5-Mev gamma; however, this value is less than that obtained with the SRE heat exchanger at a reactor power level of 10 Mw, and study of this neutron source was discontinued.

## Photon Irradiation Experiments

On the basis of simple damage theory, Oen and Holmes[63] made extensive calculations of the displacement cross sections for the pair-production and Compton-electron process of a number of materials for a variety of photon energies and lattice-atom displacement energies. These results may be used to estimate the yield of lattice defects for a given exposure of monoenergetic photons directly, or they may be used in combination with the capture-gamma-spectrum measurements of Bartholomew and Higgs[64] to calculate the defect yield expected for a photon spectrum, e.g., that from cadmium capture-gamma rays.[61]

In a study of the effect of gamma radiation on the electrical behavior of a number of semiconductors,[65] we tested these calculations experimentally. Table IV-12 lists the ratios of the calculated to the observed rate of defect introduction for several semiconducting materials for five different photon energies. The observed values were obtained from experimental measurements made at 77°K following irradiation at 40°C and were based on the assumption that two carriers were removed per lattice defect in germanium and one in silicon and gallium–arsenic. It is evident that the agreement between theory and experiment improves as the energy of the incident photon increases.

These results suggest that the simple theory of radiation damage, which neglects the ordered arrangement of atoms in a lattice, is inadequate to explain the observed results. Therefore a more sophisticated theory that takes into account the lattice effects is necessary.

Gamma rays have also proved valuable in studies of ionic crystals,[66] metals,[67] and alloys.[68]

## Neutron Irradiation Experiments

Conventional radioisotopic neutron sources,[57] e.g., $^{124}$Sb–Be (50 curies for $8 \times 10^7$ n/sec, 60-day half-life), $^{210}$Po–Be (90 curies for $2 \times 10^8$ n/sec, 138-day half-life), or $^{239}$Pu–Be (10 curies for $2 \times 10^7$ n/sec, 24,000-year half-life), cannot be used effectively for most radiation-damage experiments on solids since the actual neutron flux or total neutron dose is too low. However, such sources can be used to demonstrate radiation-damge effects in semiconducting and ionic crystals, and it can be predicted that $^{241}$Am, $^{244}$Cm, and $^{252}$Cf neutron sources will be of great interest for a wide variety of radiation-effects studies in the future.

By-product $^{241}$Am has recently been made available by the U. S. Atomic Energy Commission. Americium-241 has a 458-year half-life, a 5.44- and a 5.48-Mev alpha radiation, and an extremely weak (0.06 Mev) gamma radiation that can be readily shielded by a container made from any material of a high atomic number. Americium sources are now com-

Table IV-12 PHOTON-INDUCED LATTICE-DEFECT INTRODUCTION RATES[56]

| Radiation | Material ($n$ type) | $N_d/\phi$* Theoretical | Observed | Ratio |
|---|---|---|---|---|
| 0.667-Mev gamma from $^{137}$Cs–$^{137m}$Ba | Ge | 0.002 | $10^{-6}$ | 2000 |
| | Ga–As | 0.010 | 0.0004 | 25 |
| 1.17- (50%) and 1.33- (50%) Mev gammas from $^{60}$Co | Ge | 0.023 | 0.0006 | 38 |
| | Si | 0.026 | 0.0007 | 37 |
| | Ga–As | 0.070 | 0.014 | 5 |
| 1.7-Mev gamma from $^{124}$Sb | Ge | 0.054 | 0.0027 | 20 |
| 1.38- (50%) and 2.76- (50%) Mev gammas from $^{24}$Na | Ge | 0.074 | 0.010 | 7.4 |
| Cd (capture) gammas | Ge | 0.076 | 0.018 | 4.2 |
| | Si | 0.046 | 0.008 | 5.8 |
| | Ga–As | 0.450 | 0.320 | 1.4 |

*$N_d$ = number of defects introduced; $\phi$ = flux, photons/(cm$^2$)(sec).

mercially available with activities as high as 3.24 curies/g, and americium–beryllium neutron sources containing up to 10 g of $^{241}Am$ with outputs to $6.8 \times 10^7$ n/sec are also available.[57]

Strain and Leddicotte[69] studied the preparation, properties, and uses of $^{241}Am$ alpha, gamma, and neutron sources. Eight cylinders were fabricated; each was $1\frac{1}{16}$ in. in diameter by $1\frac{1}{4}$ in. high and contained 0.6 g (2 curies) of $^{241}Am$ in 6 g of beryllium. Four of these sources were placed around a $\frac{3}{4}$-in.-diameter sample tube, and preliminary measurements indicated a total dose rate of $7.6 \times 10^4$ n/(cm$^2$)(sec) above the 2-Mev threshold for phosphorus (300-mb cross section) and $1.9 \times 10^4$ n/(cm$^2$)(sec) above a probable 6-Mev threshold for silica (20-mb cross section).

Samples of *n* type germanium have been exposed to all eight sources at room temperature in the same position as the phosphorus and silica and indicated a total removal of conduction electrons of about $10^{12}$ per cubic centimeter after 170 hr of irradiation. It was estimated that these samples intercepted a flux of about $10^5$ n/(cm$^2$)(sec). The average rate of removal of conduction electrons was approximately 13.4 per incident neutron; and a subsequent silicon-diode neutron-spectrum detector measurement[70] indicated that the peak neutron energy was in excess of 4 Mev. Two *n* type samples of silicon were also exposed, and the results showed an average rate of removal of conduction electrons of approximately 11.5 per incident neutron. These data agree quite well with previous measurements that have involved both monoenergetic and fission-spectrum neutron-removal rates in semiconducting materials.[56]

## Discussion

It has been estimated[70] that an annular source of $AmO_2$–Be (9 cm in length by 2.5 cm in inside diameter) containing 40 g of $^{241}Am$ in 400 g of beryllium would provide a flux of $2.7 \times 10^6$ n/(cm$^2$)(sec) and that the use of americium–beryllium metal alloy might increase this value to $3.9 \times 10^6$ n/(cm$^2$)(sec). It has also been estimated that an annular source of $AmO_2$–Be containing only 0.3 g of $^{241}Am$ might be irradiated in the ORR to produce an Am–Cm–Be source that would provide a flux of $2 \times 10^7$ n/(cm$^2$)(sec). This type of neutron generator would have a half-life of 163 days and a background gamma dose rate from fission products (average energy 0.6 Mev) of $7 \times 10^4$ r/hr or $5.5 \times 10^{10}$ photons/(cm$^2$)(sec); however, the radiation-damage capability of this flux and energy of photons compared to that of neutrons would be negligible for most experiments. The completed assembly could be reirradiated when necessary for at least one additional activation.

An extremely interesting and valuable source of neutrons can be anticipated whenever $^{244}Cm$ becomes available. Recent surveys[71] indicated that significant amounts of relatively pure (free of fission products) $^{244}Cm$ could be produced at a cost of $1000 to $2000 per gram, and a program to produce 3 kg of $^{244}Cm$ was started at the Savannah River Plant in the spring of 1964. A production capability of 6 kg (1966), 18 kg (1967), and 40 kg per year through 1972 is also indicated in these surveys.

Curium-244 has a half-life of 18.4 years, and it emits both alpha (5.80 and 5.76 Mev) and weak gamma (0.043 Mev) radiations. The data of Strain and Leddicotte[69] and of Acree[70] can be used to estimate that an annular source of 0.5 g of $^{244}Cm$ in beryllium (4 cm in length by 2.5 cm in inside diameter) would provide a maximum flux of $10^6$ n/(cm$^2$)(sec) with a peak neutron energy of about 5.5 Mev. The anticipated dose rate from 0.5 g of $^{244}Cm$ is about a factor of 10 greater than that used both to study the properties of $^{241}Am$ alpha, gamma, and neutron sources[69] and to demonstrate radiation-damage effects in semiconducting materials. A 5-g source of $^{244}Cm$ would provide an estimated flux of $10^7$ n/(cm$^2$)(sec), or nearly $10^{12}$ n/(cm$^2$)(day). A 100-g source might essentially duplicate the dose-rate capabilities of the most prolific accelerator—a Cockcroft-Walton neutron generator (deuterium–tritium reaction)—without the complication of target rotation, cooling, and exchange after a total output of $10^{13}$ to $10^{14}$ n/cm$^2$. With a suitable moderator, $^{244}Cm$ could provide an extremely useful source of thermal neutrons for activation analysis. The $^9Be(\alpha,n)^{12}C$ reaction produces neutrons and 4.47-Mev gammas; however, the neutron energy, i.e., 5-Mev peak and 12-Mev maximum, is great enough that the resultant damage would exceed any gamma-ray damage for most materials.

In summary, it is evident that continued advances in radioisotope production and source development will be of extreme importance to

those who are interested in the general problem of radiation-effects studies in solids. The unique advantage of a different neutron spectrum available with radioisotopic sources and the large total-dose capabilities will provide for the first time a powerful tool for the study of disordered regions and defect clusters introduced by energetic neutrons in a wide variety of materials.

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Chapter VI. Radiation Polymerization

1. Free Radical Radiation Polymerization
   - *a.* Fundamental Principles of Radiation Polymerization by Free-Radical Mechanism
   - *b.* Dependence of the Rate of Polymerization on the Dose Rate
   - *c.* The Effect of Oxygen
   - *d.* Other Evidence in Favor of a Radical Mechanism
   - *e.* Gel Effect and After Effect
   - *f.* Comparative Rates of Polymerization of Vinyl Monomers
   - *g.* Radiation Polymerization in Solvents and in Emulsions
   - *h.* Radiation Telomerization
2. Radiation Polymerization Proceeding by an Ionic Mechanism
3. Characteristic Features of Radiation Polymerization in the Solid Phase
4. Bibliography (97 references)

Chapter VII. The Action of Ionizing Radiations on Polymers

1. Main Reactions Occurring in Polymers as the Result of Irradiation. These are (a) formation of chemical links between the macromolecular chains (cross-linking), (b) degradation and decomposition of macromolecules, evidenced by a decrease in the molecular weight, (c) changes in the concentration and the nature of the double bonds, (d) oxidative (in the presence of oxygen) and other reactions, and (e) evolution of gases.
2. Free-Radical Formation on Irradiated High Polymers. The most important are (a) formation of molecular hydrogen, (b) double-bond formation, (c) free-radical recombination, which may result in the formation of lateral bonds, branched molecules, or longer molecules, (d) exchange reactions (valency transfer between chains), and (e) migration of free valency along the chain.
3. Cross-Linking
4. Degradation
5. Oxygen Effects During Irradiation of Polymers
6. Other Radiation Chemical Effects in Polymers. These include gas evolution and changes in the concentration of double bonds.
7. Changes in Physical and Mechanical Properties of Polymers as the Result of Irradiation
8. Stability of Polymers to Radiation
9. Special Features of the Radiolysis of Solutions of Polymers
10. Preparation of Graft Copolymers by Irradiation. About 35 references are available on this subject. The methods can be classified under two headings: (a) grafting of monomer onto polymer during the process of irradiation (irradiation of polymer dipped in the monomer, or of polymer swollen with the monomer), and (b) grafting of monomer onto the preliminarily irradiated polymer.
11. Effect of Ionizing Radiation on Ion-Exchange Resins
12. Bibliography (139 references)

Chapter VIII. Radiation-Chemical Processes in Non-Polymeric Solid Substances

1. Radiolysis of Ionic Salts and of Crystallohydrates
2. Effect of Preliminary Irradiation on Topochemical Reactions
3. Effect of Irradiation on Physicochemical Processes at Interphase Boundaries
4. Radiolysis of Solid Compounds with Covalent Bonds
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Section V

Isotopes and Radiation Technology

# Safety

This section presents a short summary of the results of Reentry Flight Demonstration Number Two (RFD-2) and an evaluation of the damage to five beta sources that were recovered from a fire.

## Reentry Flight Demonstration Number Two (RFD-2)[1]

*Reentry Flight Demonstration Number Two (RFD-2), launched with a Scout rocket into an 800-mile suborbital flight, reached an apogee of about 484,000 ft, reentered at an angle of −6.081°, and reached a velocity of about 19,000 ft/sec. The nonradioactive mockup of a SNAP-9A isotope-powered generator was carried on this flight. Under the reentry conditions the SNAP generator was disintegrated by reentry heating, and the four simulated isotope capsules released from the generator were burned completely. No attempt was made to collect debris from the destruction of the generator and the capsules. Four additional capsules, released during reentry but before the generator burnup, provided additional information on the relation between capsule design and reentry burnup rate.*

If nuclear power systems[2,3] are to be used in space, they must be designed to operate so safely that no one ever will be exposed to significant amounts of radiation — either during the fabrication, handling, and operation of a system or when it is disposed of after having served its purpose.[4]

One of the most vital design criteria for a nuclear power system is its safety during reentry into the earth's atmosphere, either as a normal part of a vehicle's life cycle (orbiting satellites) or as the result of launch-vehicle malfunction. One means of obtaining safety is to use the heat generated during reentry to break up and disassemble the device, thereby promoting burnup of the nuclear materials. Winds would then disperse the radioactive residue at high altitudes, distributing it so widely that no radiation danger would exist.

To evaluate the operational safety of typical nuclear power sources, Sandia Corporation is conducting a series of full-scale reentry experiments. In the tests to date, inert materials have been used in the reactors and isotopic generators; therefore no radiation hazard exists. Reentry Flight Demonstration Number Two (RFD-2) was the second* in Sandia's series of safety flight tests of SNAP devices.

RFD-2 was conducted to study the reentry portion of a flight of an isotope-powered SNAP generator in which the safety feature, in the event of a late abort during the ascent phase or following orbital departure, was burnup and release of the isotopic fuel in micron-size particles at altitudes above 100,000 ft. The objectives of RFD-2 were

1. To investigate the disassembly design of an inert mockup of a SNAP isotope-powered generator by monitoring the following events during reentry:
   a. Destruction of the outer housing
   b. Destruction or separation of the thermoelectric or thermionic modules and insulating materials
   c. Exposure, ablation, and release of the simulated isotopic fuel
2. To trace the history of fuel-capsule exposure to reentry heat
3. To obtain information on fuel-capsule ablation by optically recording the spectral output of tracer-loaded simulated fuel capsules ejected from the reentry system early in the trajectory
4. To correlate flight-test results with analytical predictions and experimentally

*RFD-1 was a reentry flight test of an inert mockup of a SNAP-10 reactor. This test is described in Refs. 4 and 5.

measured heating rates by use of existing heat-transfer computer codes

5. To obtain information on the design of the RFD-2 reentry vehicle for use in the design of future tests

## Description of Test

RFD-2 was launched from the National Aeronautics and Space Administration (NASA) facility at Wallops Island, Va.[6] The facilities and instrumentation used in this test are described below.

*LAUNCH VEHICLE*

The launch vehicle was the first three-stage Scout to be flown by NASA. Original plans were to fly the normal four-stage version, but safety considerations arising from problems with the fourth stage dictated the use of a three-stage version. The four-stage motor was replaced by a specially built structural member called the "load adapter" so that the reentry vehicle could be attached to the Scout without extensive modifications.

*REENTRY SYSTEM*

Principal elements of the reentry system were (1) the simulated isotopic generator, (2) an external fuel-capsule experiment, and (3) a reentry vehicle with its associated subsystems, including a telemetry system.

*Generator.* The inert nuclear power source in RFD-2 was a mockup of isotope-powered SNAP generators of the SNAP-9A, IMP-mission, and SNAP-19 type,[7] designed by the Nuclear Division of Martin Co., Baltimore, Md. This configuration simulates a system designed to produce 20 watts of electrical power for an operational time in excess of 3 years.

In the operational unit, heat released by the radioactive decay of $^{238}Pu$ is converted directly to electrical energy by thermoelectric elements. The cold junctions of the thermoelectric elements are maintained at the proper operating temperature by controlled release of heat into space. The fuel is housed in a series of capsules constructed of superalloys to ensure safe containment during any handling and transportation accidents in the fabrication areas and at the launch site. The capsules, which have already been subjected to considerable safety testing, are also designed to prevent the release of radioactivity by surviving the blast overpressures and thermal forces that could be released by on-pad abort of a launching vehicle.

The dummy isotopic-generator fuel cavity held four simulated fuel capsules, each containing a different stable optical tracer element (sodium, lithium, barium, and gallium). When the generator disassembled under reentry heating, the capsules were released into the airstreams, where heating caused the exposed tracers to flare almost immediately. The flares were sighted by ground-based and airborne optical instruments to indicate the altitude at which generator disassembly occurred.

*External Fuel-Capsule Experiment.* Strapped to the outside of the generator were four simulated fuel capsules containing different optical tracer elements (thallium, silver, indium, and strontium) surrounded by simulated isotopic fuel. These capsules were ejected early in reentry, and under reentry heating the simulated fuel ablated, exposing the tracers. The flares from the heated tracers were recorded by optical instruments to provide information on fuel-ablation rates.

*Reentry Vehicle.* The principal functions of the reentry vehicle were (1) to provide an aerodynamically stable support for the simulated generator and the external fuel-capsule experiment, (2) to give thermal protection to reentry-system components, and (3) to provide for retardation of the reentry system just before water impact, for flotation after impact, and for retrieval by search craft.

Structurally the reentry vehicle (Fig. V-1) consisted of an aluminum shell surrounded by a smooth, concentric lay-up of phenolic-resin-impregnated Fiberglas, which was covered in turn by an ablation sheath. The load adapter, which replaced the fourth-stage Scout motor, fit into a cavity in the rear of the vehicle and served as the structural link between it and the Scout; it was ejected just before the start of reentry.

The RFD-2 telemetry system was designed to transmit information on the temperature and disassembly of the generator and on orientation and temperature of the reentry system. It included two FM/FM transmitters and an onboard tape recorder. The tape recorder stored information on events occurring during radio-frequency blackout; this information was trans-

mitted after blackout ended. The telemetry system also contained a C-band beacon to aid in radar tracking. A timer in the telemetry system initiated such in-flight functions as de-spin and ejection of the external fuel capsules.

The recovery system included a parachute, two flotation bags, a dye marker, flashing lights, a continuous-wave beacon, and a search-and-recovery-and-homing (SARAH) beacon attached to the parachute.

A de-spin system strapped to the outside of the reentry system was actuated near the start of reentry to reduce the roll rate by a factor of about 10; it was ejected after use.

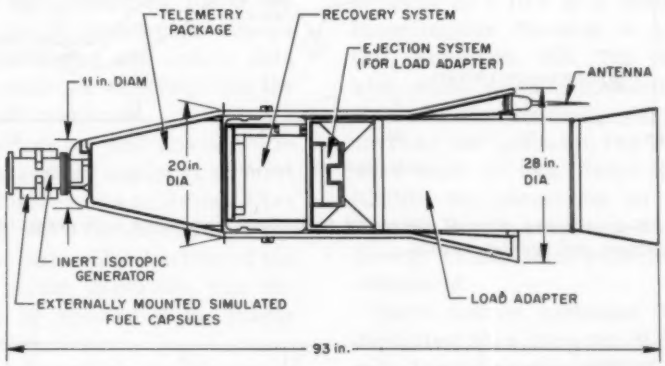


*Fig. V-1 The RFD-2 reentry vehicle (all dimensions are approximate).*

*OPTICAL INSTRUMENTATION*

Optical instrumentation included ground-based and airborne framing, plate, and spectral cameras—both fixed-axis and tracking. All ground-based optical instruments were installed at High Point, Bermuda. Sandia airborne instruments were carried in a leased DC-7B. Additional optical instruments were carried aboard two NASA DC-4's.

*RANGE INSTRUMENTATION FACILITIES*

*Wallops Island.* The NASA facility at Wallops Island provided range control, telemetry and photographic coverage of the launch, radar tracking, communications, and meteorological support.

*Bermuda.* The NASA Mercury Station at Cooper's Island furnished downrange radar and telemetry, position data for the aircraft carrying optical instruments, downrange timing, and communications. Sandia installed, maintained, and operated the optical instruments at High Point.

*Aircraft.* Seven aircraft were used to follow the reentry. These planes were equipped with optical instrumentation and telemetry-receiving and -recording equipment. The location of these planes is shown in Fig. V-2. Two aircraft from the U. S. Naval Air Station on Bermuda equipped to receive and home on the SARAH beacon were also available as search planes.

*Ships.* The NASA *Range Recoverer* and the ocean-range vessel *Sampan Hitch* (from the U. S. Air Force Missile Test Center) were stationed in the impact area. Both were equipped to receive telemetry and SARAH signals. A U. S. Coast Guard cutter was also stationed in the impact area to aid in recovery of the reentry vehicle.

## Results

RFD-2 was launched from Wallops Island just after midnight on Oct. 9, 1964. The actual trajectory (Fig. V-2) resulted in a flight path that was 20 nautical miles northeast of the predicted path (Fig. V-3) and 22 nautical miles longer. About 225 nautical miles from Wallops Island, the vehicle reached an apogee of approximately 484,000 ft and then pitched rather sharply into a reentry angle of −6.081°. Reentry velocity was about 19,000 ft/sec.

Reentry-heating and payload-disassembly events occurred as the vehicle passed south of Bermuda at a minimum ground range of 67 nautical miles. Impact occurred 8 min 20 sec after launch.

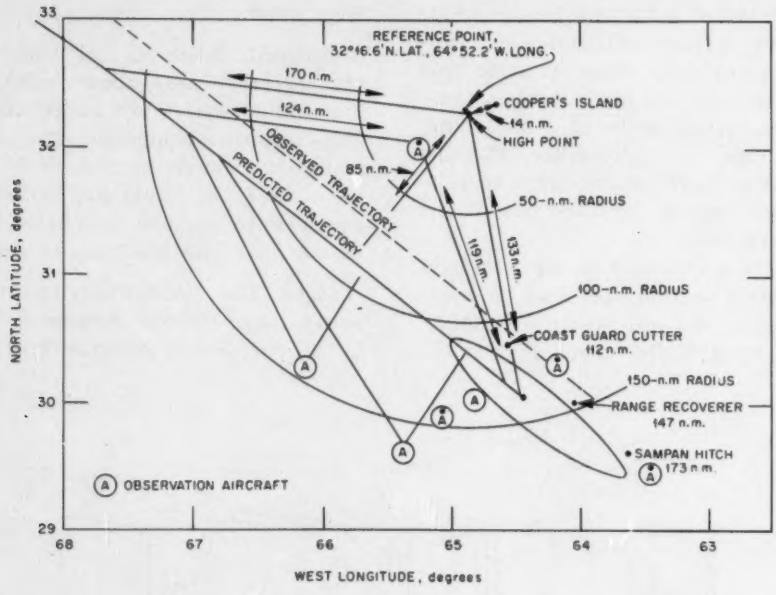


*Fig. V-2 Ground plan of observed trajectory compared with predicted trajectory and reentry events.*

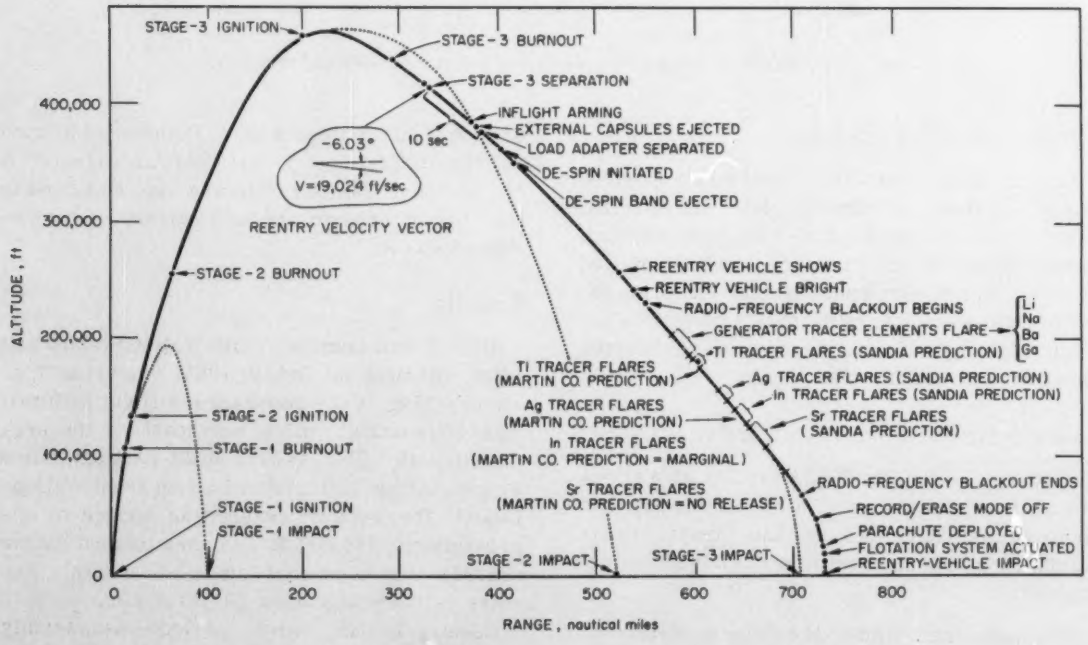


*Fig. V-3 Profile of predicted RFD-2 trajectory and associated events.*

The reentry vehicle performed satisfactorily. The recovery system functioned as prescribed by the on-board programmer in the telemetry compartment, deploying the parachute (with its attached flotation bag, light, and SARAH beacon) and inflating the internal flotation bag. However, for unknown reasons five shroud lines parted about 0.5 sec after parachute deployment. Consequently the vehicle was not recovered, although the parachute, external flotation bag, and SARAH beacon were.

High-quality telemetry data were received for the entire period of flight from lift-off to water impact; 131 of the designed 132 channels of information were returned. Optical data were recorded throughout the reentry by 108 of 111 cameras and other optical instruments covering the flight. The telemetry and optical data complemented each other in verifying that the objectives of the flight were met.

The primary objective of the mission—to demonstrate the disassembly design of an inert SNAP isotopic generator—was achieved. Disassembly and burning were verified by both the telemetry and optical data. The burning of the capsules contained in the generator was observed and verified by spectra of the tracer elements in the capsules.

The objective of determining the history of the external fuel capsules ejected early in reentry and exposed to reentry heating was met within the limits of a trajectory with a lower-than-nominal reentry velocity and a steeper-than-nominal reentry angle. Only one tracer flare (thallium) was recorded from these capsules.

The objective of correlating flight-test results with analytical predictions and experimentally measured heating rates was achieved using existing heat-transfer computer codes. Information necessary to satisfy this objective was supplied by telemetry data on temperatures at different locations in and on the generator up to the time of disassembly and by telemetry data on reentry-vehicle attitude throughout the flight.

Although the reentry vehicle was not recovered, information was obtained on its mechanical integrity. The vehicle survived and functioned properly in the environments encountered. All programmed functions seem to have occurred, and the telemetry system operated as intended throughout the flight, transmitting real-time data until the beginning of blackout and delayed-time data from the end of blackout until water impact.

# Investigation of Beta Gages Involved in Fires

By R. A. Robinson*

*Evaluation of the damage to five commercially produced beta sources that were involved in a fire showed no hazardous loss of activity as a result of the fire.*

On Nov. 11, 1964, five $^{90}Sr$ beta gages were involved in a fire at a production plant of the Bloomingdale Division of American Cyanamid Co., Aberdeen, Md. The five sources, along with some slightly contaminated rubble found close to the sources, were subsequently recovered and placed in two 30-gal drums, which were sent to Oak Ridge National Laboratory (ORNL) for inspection as part of the Safety Testing Program sponsored by the U. S. Atomic Energy Commission's Division of Isotopes Development.

Each source contained 10 to 12 mc of $^{90}Sr$ deposited on a ceramic disk. According to the manufacturer's description:

> The source compound on the ceramic disk is covered with a silicate and baked. The disk is then placed in the capsule and the capsule sealed by means of a plug which is soldered with a silver bearing solder having a melting point of 430°F. The front window, which is copper brazed to the stainless steel capsule (0.5 in. dia by 0.25 in. high), is 1.5 mils thick.

Four of the gages were in use on the first floor of the building, and the fifth gage was in use on the second floor. The fifth gage fell to the first floor during the fire when the floor collapsed. The temperatures to which the gages were exposed during the fire are not known but are estimated at between 1200 and 2000°F. The lower temperature of this range is slightly above the melting point of common cast-aluminum alloys. This estimate was based on the fact that the cast-aluminum housings in which the source capsules were mounted were completely melted away in four cases and had melted and resolidified around the source in

*Oak Ridge National Laboratory, Oak Ridge, Tenn.

the other case. The upper value of the range was based on the fact that the steel capsules, housing covers, and small screws found in the debris showed no signs of melting and there was no significant distortion. In addition, four of the copper-wire-wound solenoids that controlled the gage shutters were found in the debris sent to ORNL, and the copper, while badly oxidized, did not appear to have melted.

Visual examination of four of the sources with a stereoscopic microscope showed that the solder used for the capsule plugs had melted but had completely run out in only one source. The appearance of the source windows was:

| Source | Window |
|---|---|
| 1 | 2 small punctures, small tear at edge |
| 2 | 20% of window missing |
| 3 | Wrinkled |
| 4 | Wrinkled |

Vacuum leak tests on Nos. 3 and 4 showed a leak in No. 4 but none in No. 3. All smear tests showed contamination, and six random samples of the debris showed contamination ranging from $10^5$ to $10^7$ counts/(min)(g).

Smear tests on the fifth source, which was still in its melted aluminum housing, showed contamination levels of about 1000 counts/min. It is not known whether this was from the source itself or was cross-contamination from one of the other sources.

In the absence of information on the exact quantity of material in the original sources, it is not possible to determine the quantity of activity lost during the fire; however, direct readings of radiation from the sources indicated that only a relatively insignificant quantity in terms of the total activity could have been lost. This is further substantiated by the lack of general contamination at the fire scene. Fire equipment at the fire included radiation-survey instruments, and no contamination was detected in the area or on hoses and clothing. Representatives of the source manufacturer also made an independent survey after the fire and found no contaminated debris other than that in the two 30-gal drums sent to ORNL.

This evaluation lends support to the idea that even very lightly encapsulated sources such as beta gages can be designed so that they can undergo a reasonably typical fire situation with minimal side effects.

## References

### Reentry Flight Demonstration Number Two (RFD-2)

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2. Systems for Nuclear Auxiliary Power, A Report by the Commission, 1964, USAEC Report TID-20103, 1964. [Summarized in *Isotopes Radiation Technol.*, 1(4): 325-329 (Summer 1964).]
3. Systems for Nuclear Auxiliary Power, An Evaluation, January 1964, USAEC Report TID-20079, 1964. [Summarized in *Isotopes Radiation Technol.*, 1(4): 329-339 (Summer 1964).]
4. G. A. Fowler, Aerospace Safety of Isotopic and Reactor Power Sources, presented at the Third United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1964, Paper A/Conf.28/P/286. [Abridged version in *Isotopes Radiation Technol.*, 2(4): 350-355 (Summer 1965).]
5. R. D. Elliott, Aerospace Safety Program, SNAP-10A Reentry Flight Demonstration (RFD-1), Analysis of Flight-Test Data, USAEC Report NAA-SR-9479, Atomics International, July 15, 1964.
6. SNAP Mockup Shot to Bermuda from Virginia in Dark of Moon, *Isotopes Radiation Technol.*, 2(2): 192 (Winter 1964–1965).
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Section

VI

Isotopes and Radiation Technology

# Isotopes Around the World

Because isotope developments in other countries may suggest uses to the USAEC Division of Isotopes Development contractors, summaries of pertinent foreign work will be carried in *Isotopes and Radiation Technology* from time to time. In this issue the status of radioisotope work in Greece is presented.

## Radioisotopes in Greece*

By Myron T. Kelley†

The 1-Mw reactor at the Democritus Nuclear Research Center in Athens produces $^{131}I$, $^{82}Br$, $^{24}Na$, $^{42}K$, $^{32}P$, $^{192}Ir$, $^{51}Cr$, $^{90}Y$, and $^{198}Au$. These are used at the Center in its research work and are distributed to various hospitals and other institutions in Greece.

### Tracer Studies

Research on underground waters (see, for example, Ref. 1) is extremely important in Greece because of the scarcity of water and its increasing demand. Various experiments have been carried out at the Democritus Center in collaboration with the Institute of Geology and Subsoil Research.

Laboratory work has been performed to investigate the absorption and ion-exchange properties of tritium, chlorine, iodine, bromine, chromium, scandium, manganese, indium, and other elements used as water tracers during their passage through various geological formations. A hydrological study of the Tripolis region has been carried out with tritium as a tracer. This work was done in collaboration with the Institute of Geology and Subsoil Research and with the International Atomic Energy Agency.

A method using dead tracers and activation analysis in groundwater tracing was tested and applied to the flow of underground water in southern Greece, where the study of hydrological characteristics was the scope of the experiment as a whole. Suitable isotopes are being sought for tracing the underground waters of the Peloponnesus.

A pollution study using radioisotopes was completed at Phaleron Beach, close to the city of Athens. This study was made at a point where the main sewage canal is located, and the data will be used in preventing pollution of populated areas.

### Radioactivation Analysis

Radioactivation-analysis methods have been developed for determining arsenic, cobalt, molybdenum, copper, manganese, and zinc in plants. These elements are present in microgram or submicrogram quantities in plants and may influence their growth. This research was carried out in collaboration with the Soil and Science Laboratory of Democritus and was partially sponsored by the U. S. Department of Agriculture. Eighteen trace elements, which penetrate into aquatic organisms, have been determined in 11 Greek lakes (see, for example, Ref. 2) by activation analysis, and the same technique has been used for determining five trace elements in marine organisms. This information may help improve fish production.

Radioactivation methods have also been developed and used for determining various elements in industrial products at the request of the companies that manufacture the products. Methods were found and used for the simultaneous determination of sulfur and phosphorus and for the determination of chlorine in beer and in paper.

*Prepared from the Democritus progress report *Pepragmena*, NRC/D/CHEM-8, June 1964 (in Greek).

†Analytical Chemistry Division, Oak Ridge National Laboratory, Oak Ridge, Tenn.

Arsenic was determined by activation analysis in percussion caps and primers of several types. A new method was also developed and used for determining nickel in hydrogenated oils and fats. A radioactivation-analysis method was found for determining copper in germanium crystals.

Radioactivation analysis was combined with ion exchange to give a method for simultaneously separating and determining manganese, copper, and zinc in pine needles.

Finally, a radioactivation-analysis method has been developed for the simultaneous determination of iodine and bromine in biological materials.

## Radiography and Gaging

Radiographic control of metal manufacture and welding has been carried out for several companies.

Liquid levels and the corrosion of metals by various materials have been measured with radioisotope gaging techniques, and autoradiographic studies of chromatograms have been carried out.

## Radiation Chemistry

Two $^{60}Co$ sources of 550 and 92 curies, respectively, are in use and a third one of 3600 curies is expected to be available soon at the Center. These gamma-irradiation facilities have been used in research work over the last few years in the field of the radiation chemistry of aqueous solutions in general and of aqueous solutions of organic dyes in particular. Work already published contributes to the understanding of the specific reactions of the reactive intermediates produced in radiolysis of water with these dissolved substances. Research is in progress on the basic problem of absolute radical yields in irradiated systems and on the rate constants of reactions of the OH radical with organic and inorganic substances.

## References

### Radioisotopes in Greece

1. D. J. Burdon, E. Eriksson, T. Papadimitropoulos, N. Papakis, and B. R. Payne. The Use of Tritium in Tracing Karst Groundwater in Greece, in *Radioisotopes in Hydrology*, Symposium Proceedings, Tokyo, 1963, pp. 309-320, International Atomic Energy Agency, Vienna, 1963 (STI/PUB/71).
2. A. P. Grimanis, G. Pantazis, C. Papadopoulos, and N. Tsanos, Determination of Trace Elements in the Greek Lakes by Neutron Activation Analysis, presented at the Third United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1964, Paper A/Conf.28/P/854. [Summarized in *Isotopes Radiation Technol.*,2(4): 345-348 (Summer 1965)].

Section

VII

Isotopes and Radiation Technology

# Miscellaneous Items of Interest

## Conferences

### American Nuclear Society National Topical Meeting: "Large-Scale Production and Application of Radioisotopes"

The Savannah River Section of the Isotopes and Radiation Division of the American Nuclear Society will sponsor a national technical meeting on "Large-Scale Production and Application of Radioisotopes" in Augusta, Ga., Mar. 20–23, 1966. The term "large-scale" is defined for this meeting as involving the use of (1) more than 50,000 curies of $^{60}Co$ or other gamma emitters, (2) more than 1 kw of isotopic heat, or (3) a number of smaller units totaling in this range. "Large-scale production" will mean at least ten times these quantities per year.

Twenty-four speakers and session chairmen have accepted. The meeting is unique in that each invited speaker has agreed to act as a rapporteur and to include material submitted by others in his oral presentation. Scientists who would like their work presented should submit written reports by Jan. 2, 1966, to the program chairman:

Dr. E. J. Hennelly
Savannah River Laboratory
Aiken, S. C. 29802

The tentative technical-meeting agenda is as follows:

*Monday, March 21, AM*

1. Large-Scale Production of Radioisotopes in Production Reactors
   W. P. Overbeck, SRL
2. Large-Scale Production of Radioisotopes in Power Reactors
   A. F. Rupp, ORNL
3. Large-Scale Production of Radioisotopes in Commercial Reactors
   T. J. Slosek, Vallecitos Atomic Laboratory

*Monday, March 21, PM*

1. Large-Scale Processing and Source Preparation of Neutron-Produced Isotopes
   Eugene Lamb, ORNL
2. Large-Scale Processing and Source Preparation of Separated Fission Products
   T. S. Weissmann, Isochem, Inc.
3. Chemonuclear and Radiation Chemical Processes
   Meyer Steinberg, BNL

*Tuesday, March 22, AM*

Large-Scale Production and Application of Radioisotopes

1. Canada
   Roy F. Errington, AECL
2. Great Britain
   Harold Sheard, UKAEA
3. France and other continental European programs
   Charles Fisher, CEA, Saclay
4. Japan
   Eiji Shikata, JAERI

*Tuesday, March 22, PM*

Large-Scale Application of Radiation from Radioisotopes

1. Food Preservation
   E. S. Josephson, U. S. Army Natick Laboratories
2. New Materials
   D. E. Harmer, Dow Chemical Co.
3. Sterilization
   Charles Artandi, Ethicon, Inc.
4. Radiography
   E. W. Coleman, Picker X-Ray Corp.
5. Teletherapy
   H. D. Bruner, M. D., USAEC, Division of Biology and Medicine

*Wednesday, March 23, AM*

1. AEC Isotopic Power Applications
   Arnold Berman, USAEC, Division of Isotopes Development
2. Projected Large-Scale Use of Radioisotopes by NASA
   Robert English, NASA–Lewis
3. Long-Range Speculations on Isotopic Power
   B. I. Spinrad, ANL

4. Aerospace Nuclear Safety
   V. E. Blake, Sandia
5. Terrestrial Safety
   Alexander Aikens, Jr., USAEC, Division of Materials Licensing

### Low Energy Symposium Proceedings Now Available

The proceedings of the Symposium on Low Energy X and Gamma Sources and Applications sponsored by the AEC Division of Isotopes Development—Illinois Institute of Technology Research Institute, October 1964, in Chicago, Ill., are being printed. The proceedings should be available from CFSTI* after November 1 for about $4. [For a description of the conference, see *Isotopes and Radiation Technology*, 2(2): 190 (Winter 1964–1965.)]

Ask for the following report: Philip S. Baker and Martha Gerrard (Eds.), Symposium Proceedings, Low Energy X and Gamma Sources and Applications, Chicago, Ill., 1964, USAEC Report ORNL-IIC-5, Oak Ridge National Laboratory.

## AEC Activities

### Growing Demand for Cobalt-60 Stimulates Increased Production by AEC

The Atomic Energy Commission (AEC) is currently producing over 6 million curies of $^{60}Co$ (with an additional 3 million curies on order for future production) at its Savannah River (S. C.) Plant, with a more limited production at its Richland (Washington) Plant. The present production will be used as follows:

—0.975 million curies for studies into the characteristics and applications of high-specific-activity cobalt (300 to 600 curies/g) by Oak Ridge, Brookhaven, and Savannah River.

—1.25 million curies for reloading the irradiator at the U. S. Army Radiation Laboratory at Natick, Mass.

—0.7 million curies for studies by Savannah River of cobalt heat sources in the range of 4,000 to 10,000 thermal watts.

—0.16 million curies for the AEC's Argonne National Laboratory for use in radiation chemistry research.

—1.5 million curies for redistribution by ORNL to commercial users.

—1.44 million curies for research irradiations by Brookhaven National Laboratory.

The AEC policy is to distribute radioisotopes to commercial users only when domestic commercial producers cannot meet requirements under reasonable conditions of time, quality of product, or price.

### AEC Simplifies Regulations for Some Medical Uses of Radioisotopes

Under an amendment to the AEC regulation in the *Code of Federal Regulations*, Title 10, Part 30—Licensing of Byproduct Material—a general license is established for the use of $^{131}I$ in the form of sodium iodide for measurement of thyroid uptake; $^{125}I$ and $^{131}I$ as iodinated human serum albumin for determination of blood and blood-plasma volume; $^{58}Co$ and $^{60}Co$ for measurement of intestinal absorption of cyanocobalamin; and $^{51}Cr$ as sodium radiochromate for determination of red-blood-cell volumes and survival time. These are the only chemical forms and uses authorized, and usage is limited to physicians licensed by a state or territory to dispense drugs in the practice of medicine. The general license also limits the quantities that the licensee may possess.

Manufacture of the radiopharmaceuticals still requires a specific license from the Commission.

### Oregon and Tennessee Become the Tenth and Eleventh States To Assume Part of AEC's Regulatory Authority

Oregon and Tennessee have assumed part of the AEC's regulatory authority over the uses of radioactive materials with Commission approval. The transfer of regulatory responsibility includes licensing, rule making, and enforcement of the uses of radioisotopes, the source materials uranium and thorium, and small quantities of fissionable materials. The Oregon agreement became effective July 1, 1965, and the Tennessee agreement, Sept. 1, 1965.

The Oregon State Board of Health has been designated as the agency responsible for carrying out Oregon's radiation-control program, with the help of an appointed eight-member Radiation Advisory Committee. There are approximately 130 AEC licenses in Oregon.

*Clearinghouse for Federal Scientific and Technical Information, National Bureau of Standards, U. S. Department of Commerce, Springfield, Va. 22151.

In Tennessee the Radiological Health Service of the Tennessee Department of Public Health will conduct the radiation-control program. There are approximately 220 AEC licenses in Tennessee.

The Commission now has agreements with eleven states: Arkansas, California, Florida, Kansas, Kentucky, Mississippi, New York, North Carolina, Oregon, Tennessee, and Texas.

## Ice-Detection Device Given General License and General License Is Modified

A general license, which requires no application to the Commission, has been provided for the possession and use of 50 μc of $^{90}Sr$ in ice-detection devices as a result of a request from United Control Corp. (UCC), Redmond, Wash. For several years UCC has distributed such a device under specific license for use on aircraft.

The device operates on the same principle as other beta density gages. An electron beam passes along a sensing surface and strikes a radiation counter. Any ice on the sensing surface absorbs a portion of the beam, and the reduced signal from the radiation counter is used to control the aircraft's deicing equipment. These devices may also be used at remote or automatic ground installations.

The amendments require that the ice-detection devices be manufactured or imported according to a specific license issued by the Commission or by a state that has entered into an agreement with the Commission. The specific license, among other things, requires that the strontium be contained in such a manner that it cannot be released.

The AEC also is changing the labeling and installation requirements for general-license radioisotope devices that are routinely used as measuring, gaging, and controlling equipment or for producing light or an ionized atmosphere. The amendments result from requests by nationwide distributors of devices that the Commission and the state regulatory groups adopt uniform labeling requirements. The specific wording required on devices under Commission regulations was not acceptable under certain state regulations; hence representatives of those states and the Commission have agreed on the concept of a universal label.

Installation requirements have been clarified to state specifically when specially trained persons are required to install a radioisotope device. This requirement will be specified in the labels affixed to the device.

The amendments, effective as of late September 1965, were to Part 31, General Licenses for Certain Quantities of Byproduct Material and Byproduct Material Contained in Certain Items, and to Part 32, Specific Licenses to Manufacture, Distribute, or Import Exempted and Generally Licensed Items Containing Byproduct Material.

## Bureau of Reclamation and AEC To Develop Radioisotopic Turbine-Rating Systems

The AEC and the Bureau of Reclamation will conduct a joint three-year $300,000 development program for measuring the flow of water through high-head turbines and other hydraulic machines. Magin and Bizzell reviewed gaging of flow through turbines with the use of radioisotopes, together with applications of radioisotope technology to water-resources investigations and utilization, in an earlier issue [*Isotopes and Radiation Technology*, 2(2): 124-133 (Winter 1964–1965).]

Hydraulic engineers determine the exact volumes of water flowing at high velocities through turbines and other machinery to properly design and maintain equipment at maximum operating efficiency. Current methods of measuring flow through high-head turbines are both expensive and cumbersome and require shutting down the equipment for at least two days. In addition, the results obtained are of questionable accuracy.

Radioisotopes are already being used successfully to measure the flow of water in open channels, and hopefully this method can be adapted to closed, high-head systems.

High-head turbines are defined for this program as those with an inlet more than 100 ft above the machines. Most Bureau of Reclamation turbines are in this category, the highest being the Upper Molina in Colorado, an impulse type project with a maximum head of 2688 ft. Many other turbines, both public and private, have high heads.

Goals of the program to be undertaken include determining the best methods for injecting the radioactive tracer into flowing water above its point of entrance into high-head turbines, and developing various feedback procedures and

equipment to assure accurate control of the process and measurement of the results.

A team of hydraulic engineers and physicists in the Research Division of Reclamation's Design and Research Center in Denver, Colo., will conduct the program under the direction of Chief Engineer B. P. Bellport and Chief Research Scientist Graydon E. Burnett. The AEC will contribute to the development of the nuclear technology through its Division of Isotopes Development.

The program will be coordinated with research in universities and with facilities of national manufacturing concerns. Field tests will be conducted in Colorado, where Reclamation operates high-head turbines which are ideally suited for such tests. Final methods and equipment will be subject to acceptance as standard by the Test Code Committee of the American Society of Mechanical Engineers.

## AEC Invites Industry To Join Studies of Pasteurized Seafood at MPDI Facility

The AEC is seeking the cooperation of the seafood industry in carrying out studies of radiation-processed seafood and other marine products in the Marine Products Development Irradiator (MPDI) located at the Gloucester, Mass., Technological Laboratory of the Bureau of Commercial Fisheries. The MPDI, dedicated on Sept. 28, 1964, was described in *Isotopes and Radiation Technology*, 1(4): 310-317 (Summer 1964).

The MPDI will be available to the seafood industry for processing limited quantities of seafood for shipping, storage, and acceptability tests and for evaluation of the product in commercial plant and laboratory facilities. However, the seafoods will not be available for commercial use or for sale to the public now. Commercial seafood processors and distributors interested in negotiating a cooperative agreement should communicate with the Director, Division of Isotopes Development, U. S. Atomic Energy Commission, Washington, D. C. 20545.

## Commercial Use of Atomic Power on Oil and Gas Platform Demonstrated for the First Time in the Gulf of Mexico

Navigational aids on an offshore oil and gas platform began unattended operation on June 21, 1965, in the Gulf of Mexico. The SNAP-7F 60-watt generator produces electrical power from the radioactive decay of 14 lb of strontium titanate, with heat from the decay being converted directly to electricity by 120 pairs of thermocouples.

Located in the Gulf about 81 miles southwest of Morgan City, La., the platform is owned jointly by Phillips Petroleum Company, Bartlesville, Okla., and Kerr–McGhee Oil Company, Oklahoma City, Okla. Phillips' participation is in behalf of the Offshore Operators Committee, a trade association representing all offshore oil and gas producers operating in the Gulf of Mexico.

The nuclear generator, 22 in. in diameter and 34 in. high, is installed in one of four tubular steel legs of the platform and powers two flashing-light beacons and an electronic foghorn.

Part of the generator was fabricated at the AEC's Y-12 plant in Oak Ridge. The data obtained from the government–industry cooperative venture will be made available to all members of the offshore oil and gas industry.

## World's First Nuclear-Powered Lighthouse in Second Year of Operation

A SNAP-7B isotopic power generator has been providing the power for an unmanned beacon in the U. S. Coast Guard's Baltimore Lighthouse in Chesapeake Bay, Md., since May 20, 1964. The 60-watt nuclear generator, fueled with about 20 lb of strontium titanate and designed to operate unattended for 10 years, is 34½ in. high and 22 in. in diameter.

Inside the generator, 120 pairs of lead telluride thermocouples convert heat from the decaying fuel into electricity. Since no moving parts are involved in the generator, chances of mechanical failure are eliminated. The generator was built by the nuclear division of the Martin Co., Baltimore, Md.

## AEC Studies Ways To Increase Isotope and Radiation Use

A study was conducted for the AEC by Lauchlin M. Currie, Chairman of the AEC Advisory Committee on Isotopes and Radiation Development, and O. M. Bizzell, Chief of the Technology Utilization Branch of AEC's Division of Isotopes Development, to obtain (among

other things) recommendations as to how the AEC might better advance industrial uses of isotopes and radiation. Since much of the information obtained was proprietary in nature, the study may not be made public. In general, the 40 firms recommended that the Commission

1. Actively seek joint funding projects for isotopes and radiation developments with industry.
2. Set up procedures for loan or rental of large radiation sources.
3. Broaden AEC patent procedures.
4. Extend the use of the general licensing provision of AEC regulations in light of past experience.
5. Adopt a more positive public information program to offset unfounded fears about isotopes and radiation.

Thirty of the firms reported increasing uses of isotopes, and the Commission is studying the report to determine how the findings can be translated into an improved policy.

# Availability of Isotopes and Services

## Prices on Four Radioisotopes To Be Reduced Sharply by AEC

The AEC is making sharp price reductions on the fission products $^{90}Sr$, $^{147}Pm$, $^{144}Ce$, and $^{137}Cs$, with the new price schedule to take effect Nov. 10, 1965, on the basis of a contract with Isochem, Inc., to privately finance construction and operation of a new Fission Products Conversion and Encapsulation plant (FPCE) at the AEC site at Richland, Wash. At present the AEC's ORNL and the Martin Co.'s Quehanna, Pa., facility process these isotopes.

The FPCE, to begin operation in the fall of 1968, will be designed for large-scale production and distribution of these radioisotopes at low unit prices, opening the way for greatly expanded uses of these materials. The new lower prices are set at levels approaching the prices expected to be charged by Isochem, in order to further stimulate the market for these materials in the period before the proposed new plant begins operation.

Under the agreement being negotiated with Isochem, a joint venture of Martin Marietta Corporation and United States Rubber Company, the corporation will build the FPCE (at an estimated cost of $8 million) and operate the facility on a commercial basis. In addition, Isochem will also manage the AEC's chemical separation and related facilities at Richland under a cost-plus-fixed-fee contract. [For further information on the FPCE, see "Hanford Isotopes Plant" by C. A. Rohrmann, *Isotopes and Radiation Technology*, 2(2): 99-123 (Winter 1964–1965).] However, until the Isochem facility becomes operational in the fall of 1968, all orders should continue to be placed with ORNL.

The new prices will be applicable to all purchasers, foreign and domestic, including federal agencies. The reduced prices are expected to stimulate large-scale utilization of the radioisotopes and to help develop a source of fabricated and encapsulated fission products independent of the AEC.

The old and new price schedules are shown in Table VII-1.

Table VII-1 OLD AND NEW PRICE SCHEDULES FOR RADIOISOTOPES

| Old schedule | | New schedule | |
|---|---|---|---|
| Isotope quantity, curies | Price, $/curie | Isotope quantity, curies | Price, $/curie |
| | $^{90}Sr$ | | |
| 0–5,000 | 2.00 | 0–5,000 | 0.50 |
| 5,001–30,000 | 1.00 | 5,001–50,000 | 0.30 |
| >30,000 | 0.75 | 50,001–150,000 | 0.25 |
| | | >150,000 | 0.20 |
| | $^{147}Pm$ | | |
| 0–20,000 | 5.00 | 0–5,000 | 0.50 |
| 20,001–50,000 | 2.50 | 5,001–50,000 | 0.30 |
| >50,000 | 1.75 | 50,001–150,000 | 0.25 |
| | | >150,000 | 0.20 |
| | $^{144}Ce$ | | |
| 0–20,000 | 2.00 | 0–5,000 | 0.50 |
| 20,001–100,000 | 1.50 | 5,001–50,000 | 0.30 |
| >100,000 | 1.00 | 50,001–150,000 | 0.20 |
| | | >150,000 | 0.15 |
| | $^{137}Cs$ | | |
| 0–20,000 | 1.00 | 0–10,000 | 0.50 |
| 20,001–100,000 | 0.75 | 10,001–50,000 | 0.45 |
| >100,000 | 0.50 | 50,001–200,000 | 0.35 |
| | | >200,000 | 0.125 |

## High-Purity Stable Isotopes Available from ORNL

Recent additions of high-purity electromagnetically separated isotopes to the ORNL stable-isotopes inventory include:

| Isotope | Purity, % | Amount available, g |
|---|---|---|
| $^{30}Si$ | 91.0 | 0.83 |
| $^{32}S$ | 99.8 | 13.0 |
| $^{33}S$ | 61.1 | 0.28 |
| $^{34}S$ | 85.6 | 0.69 |
| $^{69}Ga$ | 99.7 | 21.26 |
| $^{71}Ga$ | 99.6 | 9.29 |
| $^{74}Se$ | 59.1 | 0.30 |
| $^{96}Ru$ | 98.0 | 0.70 |
| $^{99}Ru$ | 98.2 | 4.49 |
| $^{101}Ru$ | 97.7 | 1.70 |
| $^{102}Ru$ | 99.5 | 2.40 |
| $^{104}Ru$ | 99.7 | 2.00 |
| $^{123}Te$ | 83.8 | 0.62 |
| $^{196}Hg$ | 45.3 | 0.026 |

### Nuclear Science and Engineering Corporation Improves Line

Nuclear Science and Engineering Corporation (NSEC) of Pittsburgh, Pa., recently announced its expanded line of radioisotopes. Since February 1965, 39 radioisotopes have been added to NSEC's catalog to bring their total to 73. Among the new products are 3 popular tracers ($^{131}I$, $^{32}P$, and $^{35}S$), 5 rare-earth isotopes, and 14 short-lived radioisotopes (including $^{198}Au$, $^{24}Na$, $^{197}Hg$, $^{42}K$, $^{64}Cu$, and $^{82}Br$). Prices of the popular tracers and the 14 short-lived radioisotopes are reported to be below those of the AEC.

The AEC will continue to supply those isotopes not commercially available, as well as materials meeting higher technical specifications than those commercially available.

## General

### ORNL Makes Largest Commercial Shipment of Radioactive Material to France

The largest single commercial shipment of radioactive material to date was made in April 1965 by the Isotopes Division of Oak Ridge National Laboratory. The shipment consisted of 173,520 curies of cesium chloride for use in an irradiation facility in France. ORNL has made larger shipments of radioactive cesium and strontium, but these earlier orders were for use in AEC-sponsored programs rather than for commercial users.

This shipment went to Saint-Gobain Techniques Nouvelles in Seine, France, and was installed in a trailer-mounted irradiator for use in a broad industrial and commercial radiation program that includes food irradiation. The cesium shipment, weighing just under 15 lb, was loaded in a steel and lead container weighing over 7 tons and measuring 4 ft in diameter by 5½ ft high. ORNL fabricated the $^{137}Cs$ in pellet form and encapsulated it in twenty 1- by 12-in. stainless-steel containers.

### New Research Reactor Begins Operation at Oak Ridge

The High Flux Isotope Reactor (HFIR) at ORNL went critical on Aug. 25, 1965. Operation of the HFIR has the objective of making available relatively large quantities of transplutonium elements for research and development. Production of berkelium, californium, einsteinium, and fermium from HFIR is expected to be in milligram to gram amounts.

The transplutonium elements will be used in basic research into nuclear structure and spontaneous fission, decay processes, and heavy-element chemistry. Scientists also hope to gather information that will help explain natural phenomena such as the formation of stellar systems.

The HFIR, with a 9.4-kg 93% $^{235}U$ core, has a design power of 100 Mw(t). The high power density in the fuel core will require that the fuel be replaced about every 2 weeks.

The transplutonium-element production cycle began in 1959 with the production of 300 g of $^{242}Pu$ at the Commission's Savannah River Plant. After suitable irradiation in the HFIR, the targets will be removed and the curium separated. The curium will then be prepared for further irradiation. After an extended number of such cycles, annual recoveries are predicted of gram amounts of californium, hundreds of milligrams of berkelium, tens of milligrams of einsteinium, and about a milligram of fermium. About 100 g of the heavier isotopes of curium will also be recovered.

Although the HFIR's primary purpose is the production of transplutonium elements, the reactor has several experimental facilities, including hydraulic tubes designated for production of isotopes of the lighter elements. Because of the extremely high flux, the reactor will produce many radioisotopes with higher specific activities and yields than now feasible, as well as certain long-lived activities which cannot be

economically produced at present because of low cross sections of the target nuclides. Studies will include the possible production of radioisotopes by secondary neutron capture, increasing specific activities by target atom "burnup" processes, and the effect of secondary reactions on the purity of radioisotopes produced routinely. Another interesting possible use of the HFIR is the enrichment of certain stable isotopes by burnup of the unwanted nuclides.

The reactor is one of three facilities at ORNL devoted to the transplutonium program. The other two are the Transuranium Processing Plant and the Transuranium Research Laboratory.

### Use of Cesium Reported in USSR

The USSR is using $^{137}Cs$ instead of $^{60}Co$ in a gamma teletherapy unit. Radiologists at a Moscow hospital credited the longer half-life of cesium for the change. No further details were given. ORNL has recently experienced an increase in sales of $^{137}Cs$ teletherapy sources, particularly to Germany and Italy.

(See page 80, this issue, for report on ORNL shipment of $^{137}Cs$ to France.)

### Gemini V Uses Tracers in Space

Labeled serum albumin was injected into the bloodstreams of astronauts L. Gordon Cooper, Jr., and Charles Conrad, Jr., just before Gemini V launch. Each man received 200 μc of $^{51}Cr$ and 7.5 μc of $^{125}I$, a dose of about 88 mr.

Under investigation is the red-blood-cell loss observed on previous long space flights. Fluid volume of circulating blood is given by $^{125}I$ and red-cell mass by $^{51}Cr$.

## Translations Available

As part of the effort to cover the foreign literature in the isotopes field, selected articles are translated by members of the ORNL Isotopes Development Center. These articles are available through the usual USAEC information dissemination channels (see front cover). Some of the recent translations of interest in the field of production and applications of isotopes are:

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2. ORNL-tr-308, J. Beydon, G. Delmas, J. Druaut, and P. Griboval, Extraction of $^{198}_{80}Hg$ Formed in Gold by Irradiation with Slow Neutrons in the Saclay P-2 Reactor, translated by Martha Gerrard and J. J. Pinajian from *J. Phys. Radium* (French), 19: Suppl. 144A-146A (1958).

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